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(November 1985)

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Inventories	Motion	Emission Inventories	15E	12A
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SESSION 1: DEVELOPMENT OF NAPAP EMISSION INVENTORIES FOR ANTHROPOGENIC SOURCES

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OVERVIEW OF NAPAP EMISSIONS INVENTORY ACTIVITIES FOR ANTHROPOGENIC SOURCES

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Charleston, South Carolina

November 12-14, 1985

OVERVIEW OF NAPAP EMISSIONS INVENTORY ACTIVITIES FOR ANTHROPOGENIC SOURCES

ABSTRACT

The National Acid Precipitation Assessment Program (NAPAP) has charged its Task Group B on Man-Made Sources with the development of comprehensive and accurate inventories of emissions from anthropogenic sources believed to be important in acid deposition processes. This effort involves developing estimates of past, present, and future acid deposition precursor emissions with adequate geographic, temporal, and sectoral resolution to support the research requirements of NAPAP. It also entails quantifying the degree of uncertainty associated with those estimates. The major anticipated needs for emissions data include: historical analyses of short- and long-term trends in emissions; atmospheric modeling using Lagrangian and Eulerian techniques; and overall assessments of the acid deposition phenomenon. This paper presents an overview of the current and future activities related to the development of anthropogenic emissions inventory data. Included are descriptions of the products that have been generated by Task Group B and plans for the integration of activities surrounding the development of the 1985 NAPAP Emissions Inventory.

OVERVIEW OF NAPAP EMISSIONS INVENTORY ACTIVITIES FOR ANTHROPOGENIC SOURCES

INTRODUCTION

The National Acid Precipitation Assessment Program (NAPAP) was established by Congress in 1980 to coordinate and expand research on problems posed by acid deposition in and around the United States. The program is composed of 10 task groups having specific technical responsibilites. One of these groups, Task Group B: Man-Made Sources, is charged with providing a complete and accurate inventory of emissions from anthropogenic sources thought to be important in acid-deposition processes.

To fulfill its stated objectives and to support other related NAPAP research, Task Group B either has generated or is planning the following major data bases:

- Historical Emissions Estimates.
- 1980 NAPAP Emissions Inventory (Versions 1.0 through 5.2).
- 1985 NAPAP Emissions Inventory.
- Regional Acid Deposition Model Field Validation Emissions Inventory.

The primary focus of FY 85 emissions inventory activities was directed toward fulfillment of the emissions data base requirements for development and testing of the Eulerian Regional Acid Deposition Model (RADM). Within the EPA's Office of Research and Development, the Atmospheric Sciences Research Laboratory has been assigned the lead responsibility for developing the RADM. National Center for Atmospheric Research (NCAR) has been assigned the task of developing the model framework which includes a number of chemical transformation modules. The variety of these modules under development dictates the specific chemical species required in the emissions inventory.

The FY 85 emissions inventory development program was also directly supportive of certain "assessment" activities. The emissions inventory's needs for assessment purposes are generally less detailed and resource intensive than modeling requirements, but are nonetheless extremely important to the overall NAPAP program. Assessment projects use more aggregated emissions information in combination with models or other analytic techniques to evaluate the relative importance of various pollutants, regions, and types of sources, to the acid deposition problem. Results of these studies have implications for planning additional research needed and development of potential control/mitigation strategies.

DISCUSSION

The major tasks accomplished during FY 85 and planned for FY 86 are discussed below.

Historical Emissions Estimates

In order to provide a basis for evaluating acid precipitation related damage and for studying trends in water chemistry and deposition monitoring data, it is important to understand both historic (long-term) and recent (short-term) trends in emissions. Historic SO_x and NO_x emission levels for the U.S. were estimated by individual source category on the state level from 1900 to 1980 for every fifth year and for 1978. These emissions were calculated from fuel consumption and industrial output statistics, estimates of average statewide fuel properties, and estimates of emission factors specific to each source category. The emissions estimates were then aggregated to show the emissions trends by state, region, and all states combined.

To support a number of short-term trends analyses, Task Group B developed monthly SO_2 and NO_x emissions estimates for the years 1975 through 1983. Annual state total SO_2 and NO_x estimates were used with fuel consumption and industrial production data to provide monthly values on a state-by-state basis. A system to provide continually updated and refined monthly data, using periodically published information, was also formulated as part of this effort.

Long- and short-term historical SO_2 emissions are depicted in Figures 1 and 2, respectively.

1980 Emissions Inventory

The contents of the 1980 NAPAP Emissions Inventory (Version 5.0) and corresponding Eulerian Modeling Inventory (Version 5.2) are shown in Figure 3. Compilation of these products required the integration of several unique, but related, work efforts, as shown in Figure 4.

The 1980 NAPAP Emissions Inventory, representing the most comprehensive and highest quality emissions data available for the 1980 base year, was completed. The emissions inventory was compiled, starting with the 1980 "snapshot" inventory, from EPA's National Emissions Data System (NEDS). Updates were made to address known deficiencies in the NEDS data. These included substitutions of specific data with information from other inventories or data bases that were believed to be more reliable (e.g., replacement of fuel data for major utility boilers using 1980 Federal Power Commission data). Emission factors and corresponding emissions estimates for a number of pollutants not contained in NEDS were also developed and incorporated. Data covering Canada were added, and improvements were made to methods used to calculate area source and highway vehicle emissions. Systematic quality control checks were also completed for all significant data items in the emissions inventory, and estimates of uncertainty were made.

The importance of the utility industry has been recognized by Task Group B, which is responsible for not only developing estimates of current emissions from the utility sector but also for projecting future emissions. Because of different but overlapping needs, Task Group B has developed a single data base that will meet the utility-related data needs of all NAPAP task groups, the NAPAP Utility Reference File (NURF). The NURF consists of four component files: the master unit data file (containing information on all electric utility generating units), the fuel specific emissions file (containing emissions information by fuel and input), the stack parameters file (containing stack information on a unit-specific basis), and the announced unit file (containing unit-specific information on units coming on-line or converting to coal-firing after 1980). The NURF was utilized to derive and verify emissions data from the utility sector in the 1980 NAPAP Emissions Inventory.

As previously stated, one of the most extensive applications of the 1980 NAPAP Emissions Inventory is to support development and testing of the RADM. For this purpose, the 1980 NAPAP Emissions Inventory (Version 5.0) had to be further resolved temporally, spatially, and by component species (Version 5.2).

Annual emissions had to be allocated to hourly profiles for a typical weekday, Saturday, and Sunday in each season; county-level area sources and minor point sources had to be spatially resolved to modeling grid cells; NO_x emissions had to be split into NO and NO_2; TSP emissions had to be assigned to alkalinity classes; and VOC emissions had to be apportioned into 30 photochemical reactivity classes.

Development of a methodology and a computerized data base for estimating the uncertainty associated with the 1980 NAPAP Emissions Inventory was also undertaken. A workshop was held during which expert opinion was solicited and a modified Delphi method was applied to arrive at quantitative estimates for the auxiliary data needed to calculate the uncertainties of emissions values. Methodologies are now being developed for the use of expert opinion to generate the auxiliary data needed to calculate emissions uncertainties for the NAPAP 1985 base year.

1985 Emissions Inventory

FY 86 activities have been focused on the development of the 1985 NAPAP Emissions Inventory. Much of the work that went into the development of the 1980 inventory will be drawn upon to create the 1985 data base, as shown in Figure 5. One important departure from the approach used to develop the 1980 NAPAP Emissions Inventory is the enlistment of individual state agencies to supply new 1985 emissions data for the NEDS system, specifically for the NAPAP effort. Previous efforts have started with NEDS "snapshot" files which did not necessarily represent the best data available from the states. As a result, a major state interface/assistance component will be involved.

In addition, efforts related to development of the 1985 data base will include: incorporation of 1985 Canadian data, 1985-specific temporal allocation factor development, enhancements to spatial allocation and speciation data, verification tests to support emission factor refinements, and an enhanced QA program.

RADM Field Evaluation Emissions Inventory

Another focus area for FY 86 relates to a proposed field study for validating the Eulerian Regional Acid Deposition Model (RADM). This would entail development of an "episodic" emissions inventory for pre-set intensive study periods including collection of actual (rather than typical) hourly emission values using Continuous Emissions Monitoring (CEM) for major sources, and accounting for phenomena such as major forest fires, chemical spills, and process upsets. The episodic inventories could then be used with corresponding deposition and atmospheric chemistry measurements to validate the RADM.

CONCLUSIONS

Task Group B has generated a number of data bases to fulfill its stated objectives. These include estimates of historic emissions back to 1900 to support long-term trends analyses; estimates of monthly emissions from 1975 through 1983 to support short-term trends analyses; estimates of 1980 emissions on both an annual basis, and with the necessary temporal, spatial, and pollutant species resolution to support Eulerian modeling analyses; and estimates of uncertainty associated with the 1980 NAPAP Emissions Inventory. Work planned for FY 86 includes development of an emissions inventory for the 1985 base year, and development of an episodic emissions inventory to support field validation of the RADM.

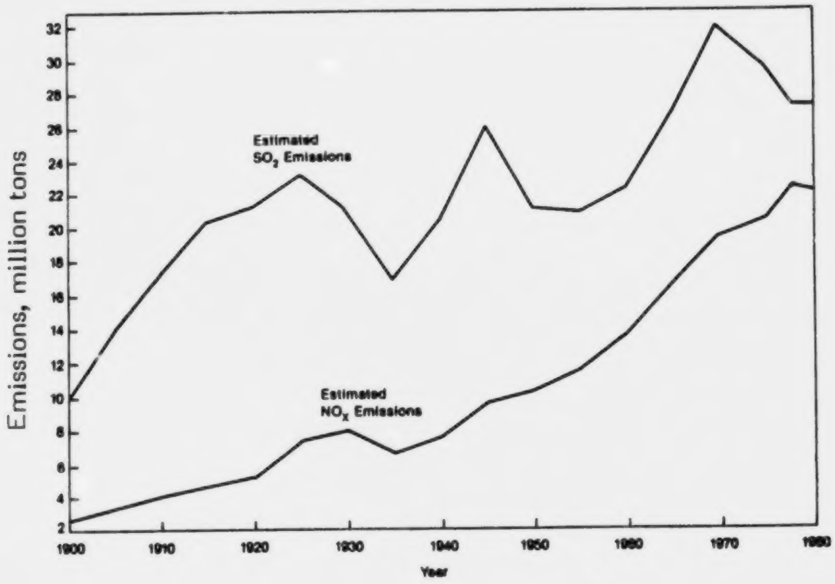

Figure 1. Overall trend in SO_2 and NO_x emissions from 1900 to 1980 for the U.S. and by source category for each study year.

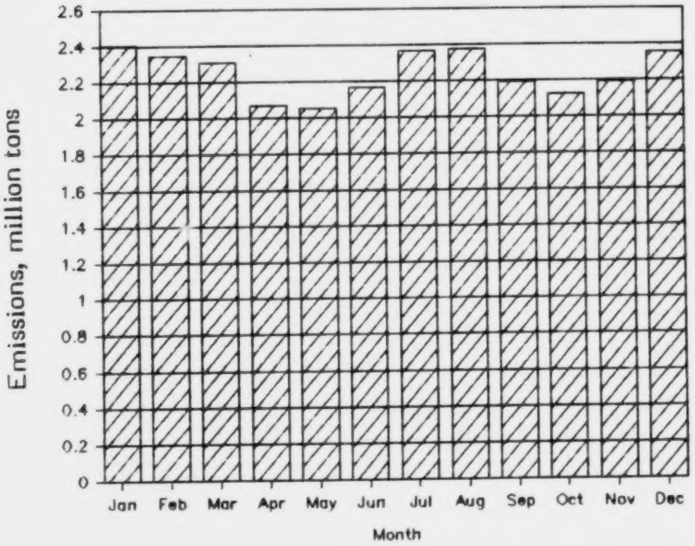

Figure 2. Monthly trends in U.S. SO_2 emissions for 1980.

	1980 NAPAP ANNUAL EMISSIONS INVENTORY	1980 NAPAP EULERIAN ACID DEPOSITION MODELING EMISSIONS INVENTORY	
TEMPORAL RESOLUTION	Annual/seasonal	Hourly emissions values for a typical weekday, Saturday, and Sunday for all four seasons	
GEOGRAPHIC DOMAIN/ RESOLUTION	48 Contiguous states and Canada with area sources at the county level	48 States and Canada; separate major point source file with minor point sources and area sources assigned to 63,000 20 x 20 km grid cells	
POLLUTANTS	SO_2	SO_2	VOC (cont.)
	SO_4	SO_4	- Isobutene
	TSP	TSP:	- Trans-2 butene
	Pb	- Ca	- Pentane
	CO	- Mg	- Isopentane
	HCl	- K	- 2,3-dimethylbutane
	HF	- Na	- Other alkenes
	NO_x	Pb	- Other alkanes
	NH_3	CO	- Formic acid
	VOC	HCl	- Acetic acid
		HF	- Other organic acids
		NO	- Formaldehyde
		NO_2	- Acetaldehyde
		NH_3	- Propionaldehyde
		VOC:	- Acetone
		- Methane	- Other ketones
		- Ethane	- Other aldehydes
		- Ethylene	- Xylene
		- Propane	- Benzene
		- Propylene	- Toluene
		- N-butane	- Ethylbenzene
		- 1,2-butene	- Other aromatics
		- Isobutane	

Figure 3. Contents of the 1980 NAPAP emissions inventories.

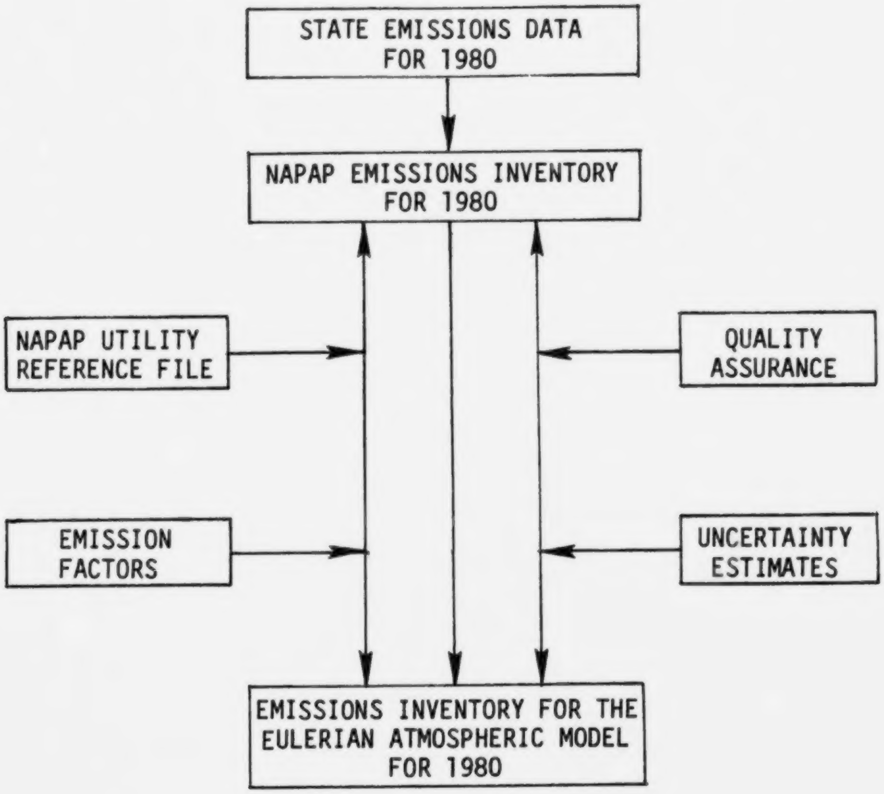
STATE EMISSIONS DATA FOR 1980
NAPAP EMISSIONS INVENTORY FOR 1980
NAPAP UTILITY REFERENCE FILE
QUALITY ASSURANCE
EMISSION FACTORS
UNCERTAINTY ESTIMATES
EMISSIONS INVENTORY FOR THE EULERIAN ATMOSPHERIC MODEL FOR 1980

Figure 4. Major program components for developing the 1980 NAPAP Emissions Inventory.

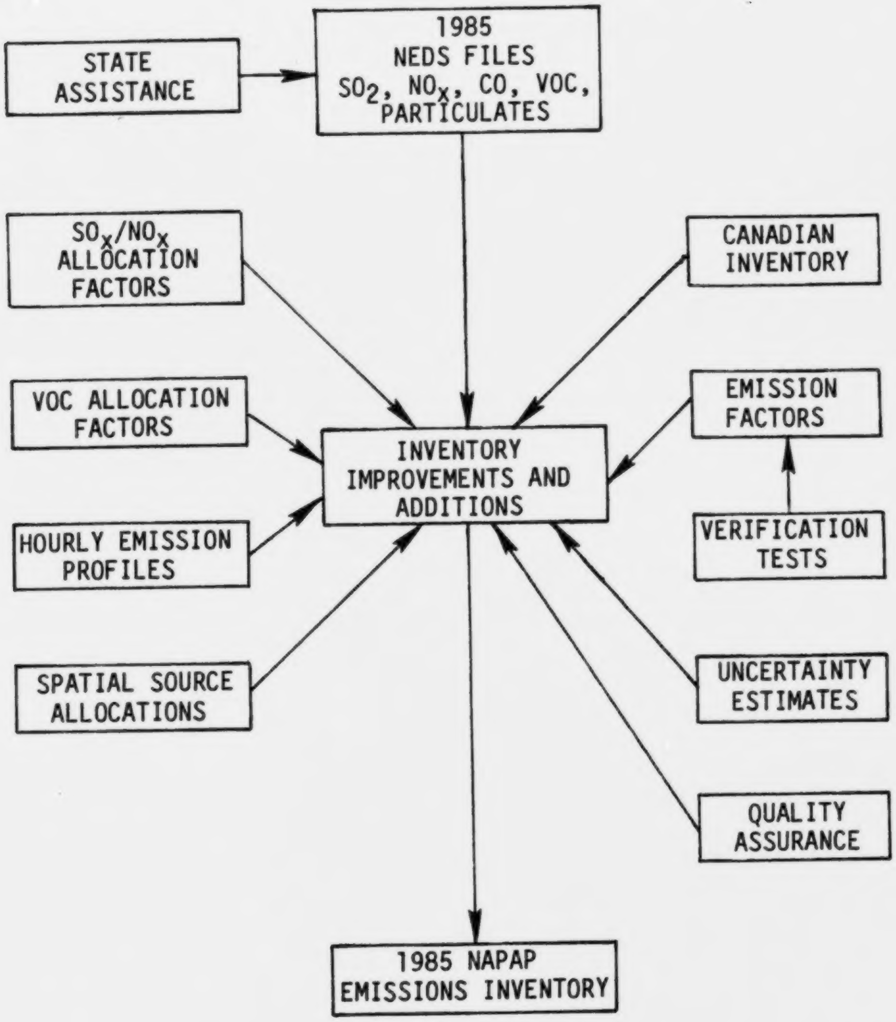
STATE ASSISTANCE
1985 NEDS FILES SO_2, NO_x, CO, VOC, PARTICULATES
SO_x/NO_x ALLOCATION FACTORS
CANADIAN INVENTORY
VOC ALLOCATION FACTORS
EMISSION FACTORS
INVENTORY IMPROVEMENTS AND ADDITIONS
HOURLY EMISSION PROFILES
VERIFICATION TESTS
SPATIAL SOURCE ALLOCATIONS
UNCERTAINTY ESTIMATES
QUALITY ASSURANCE
1985 NAPAP EMISSIONS INVENTORY

Figure 5. Integration of activities for developing the 1985 NAPAP Emissions Inventory.

ASSESSMENT OF THE FINAL
1980 NAPAP EMISSIONS INVENTORY

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November 12-14, 1985

ASSESSMENT OF THE FINAL
1980 NAPAP EMISSIONS INVENTORY

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ABSTRACT

A detailed 1980 base year emissions inventory has been developed to support the needs of various task groups within the National Acid Precipitation Assessment Program (NAPAP). The emissions inventory was compiled starting with the 1980 "snapshot" inventory from EPA's National Emissions Data System (NEDS) which was converted to Emissions Inventory System (EIS) format. Updates were made to address areas where the NEDS inventory was known to be deficient. In addition to SO_x, NO_x, non-methane VOC, TSP, and CO which are included in NEDS, emissions estimates for SO_4, NH_3, total VOC, HCl, HF, and Pb were generated. Emissions data representing 1980 conditions were obtained from Environment Canada, converted to EIS format, and incorporated into the 1980 NAPAP emissions data base. VOC emissions estimates for the highway vehicle, residential wood combustion, and organic solvent use area source categories have been improved. Systematic quality control checks have been completed for all significant data items in the emissions inventory for major point sources. The most recent version of the emissions inventory, Version 5.0, is now being finalized. Therefore, emissions data presented herein reflect Versions 3.0 and 4.0 for which detailed summaries are available. Emissions of SO_x, NO_x, and VOC, which are of primary interest for acid deposition research, are 27.1, 23.7, and 23.3 million tons per year, respectively. Emissions in the NAPAP data base agree reasonably well with Work Group 3B and EPA/OAQPS emissions trends estimates. NAPAP fuel use data show reasonable agreement with fuel values in DOE's State Energy Data Report. Version 5.0 of the NAPAP data base represents the best detailed 1980 emissions inventory presently available.

ASSESSMENT OF THE FINAL
1980 NAPAP EMISSIONS INVENTORY

INTRODUCTION

This project required development of a central, quality-assured data base of emissions of pollutants of interest for acid deposition research and modeling. The pollutants believed to be most important in acid deposition processes -- sulfur oxides, nitrogen oxides, primary sulfate, volatile organic compounds (VOCs), ammonia, chlorides, fluorides, and others -- were addressed. The data needed consist primarily of emissions estimates for the 1980 base year with adequate geographic and temporal detail. The area covered includes the 48 contiguous states of the U.S., the District of Columbia, and the 10 provinces and 2 territories of Canada.

The starting point for the 1980 NAPAP emissions data base was the 1980 "snapshot" data file of the National Emissions Data System (NEDS), maintained by the U.S. Environmental Protection Agency (EPA) at Research Triangle Park, North Carolina. Activities to create the initial detailed NAPAP emissions inventory from the 1980 NEDS "snapshot" file began in January 1983. The Emissions Inventory System (EIS) was selected to store the NAPAP emissions data base. A goal of the initial conversion to EIS was to maintain equivalency between emissions totals in NEDS and EIS.

DISCUSSION

In an effort to improve the quality of the initial NAPAP emissions data base, updates were initiated to address areas where the NEDS inventory was known to be deficient. The following updates resulted in Version 1.0 of the inventory (11/29/83) and include:

- Incorporating latest EPA emission factors (through Supplement 14 of AP-42).
- Substituting other NEDS point source data that more represent calendar year 1980 for 12 states.

- Updating fuel type, quantity and quality, source classification code (SCC), control equipment and efficiency, and boiler capacity for electric utilities that could be matched in the Pechan unit inventory (1980) and calculating updated emissions using AP-42 emission factors.
- Updating seasonal throughput for all boilers matched in the Pechan unit inventory using FPC Form 4 data for 1980.
- Adding seasonal factors for area source emissions by category.

Subsequent updates which resulted in Version 2.0 of the 1980 NAPAP emissions inventory (12/30/83) include:

- Updating copper smelters to reflect SO_x emissions in Work Group 3B inventory (1980).
- Substituting plants in Northeast Corridor Regional Modeling Project (NECRMP) inventory (1980) for those in NAPAP except for power plants updated with Pechan data and plants that exhibited no improvement in SO_x emissions.
- Incorporating through Supplement 14 emission factors in NECRMP data substituted.
- Correcting several problems with previous power plant updates.

Additional updates made to the emissions inventory in generating Version 3.0 (6/27/84) include:

- Updating power plant SO_x emissions to be consistent with Pechan plant inventory.
- Correcting NAPAP as appropriate to eliminate the largest fuel discrepancies between NAPAP and DOE's State Energy Data Report (SEDR).
- Updating area sources to incorporate data for 15 counties previously omitted.
- Making additional power plant updates for consistency with the Pechan unit inventory, to add plants/units not previously matched, and to better identify plant names.
- Revising incorrect SCCs associated with fuel discrepancies and power plant changes.

In creating Version 4.0 of the data base (9/18/84), the improvements made include:

- Adding sulfate and ammonia emission factors, control device data, and emissions.

- Eliminating roundoff error present in earlier emission factor files for point sources.
- Incorporating emissions estimates for point sources/pollutants that had an emission factor but no emission estimate in the file.
- Adding latitude/longitude and/or Universal Traverse Mercator (UTM) coordinates for all point sources.

After some additional processing to put the data base in the format needed by GCA, this version of the inventory was supplied to GCA for generating Eulerian model input.

Version 5.0, the final 1980 NAPAP emissions inventory, was scheduled for completion by October 31, 1985. The additional updates being made to create Version 5.0 of the data base include:

- Incorporating 1980 Canadian point and area source emissions data.
- Substituting improved 1980 utility data from the NAPAP Utility Reference File.
- Adding chloride, fluoride, and lead emission factors, control device data, and emissions.
- Performing a systematic quality control check for important data base items to identify missing or anomalous data and correct as appropriate using average or typical values.
- Updating area source data for highway vehicles, residential wood combustion, and organic solvent use to reflect improved data/procedures for calculating VOC emissions.
- Adding total VOC emissions to the inventory for point and area sources (previous emissions reflected non-methane VOC only).

Emissions summaries of the 1980 NAPAP emissions data base are presented in this section. Table 1 shows the difference in emissions for the five most important pollutants from the four existing versions of the data base. Little difference exists between Versions 3.0 and 4.0 for SO_x, NO_x, and VOC. Table 2 indicates that SO_x emissions are dominated by electric utilities, primarily from coal-fired generating stations in the eastern U.S. For NO_x, the largest sources are transportation (mostly highway vehicles), electric utilities, and industrial combustion. VOC emissions result largely from miscellaneous sources (including organic solvent use, gasoline service stations, and forest wildfires), transportation, and other industrial processes. The geographic breakdown of SO_x

emissions in Table 3 shows that EPA Regions 5 and 4 are the largest contributors, and the eastern 31 states account for over 82% of total emissions. Regions 6, 5, and 4 are the highest NO_x emitters, with the eastern 31 states accounting for about 64% of national emissions. Regions 5, 4, and 6 contribute the largest amount of VOC emissions, and the eastern 31 states account for about 66% of nationwide emissions. Table 4 indicates that nearly all VOC and about 68% of NO_x emissions are released below 120 feet. Nearly 40% of all SO_x is released above 480 feet. Table 5 identifies source category contributions to nationwide sulfate and ammonia emissions. Electric utilities and industrial point sources account for over 80% of the total sulfate emissions while area sources, primarily manure field application, contribute about 80% of the total ammonia emissions. Table 6 summarizes Canadian emissions by source category and indicates that electric utilities have much less influence than in the United States. Smelters and area sources, however, are more significant emitters in Canada.

CONCLUSIONS

The point and area source inventory represents year 1980 well with over 80% of emissions of SO_x, NO_x, and VOC having an indicated year of record of 1980. Virtually all significant SO_x and NO_x emitting plants are included in the NAPAP point source file. Outside of the NECRMP domain, the completeness of the NAPAP point source file for VOC is likely to be inconsistent depending on the thoroughness of the states. All point sources have location coordinates and reasonable stack data in Version 5.0.

Fuel summaries from NAPAP Version 3.0 and the SEDR show reasonable agreement with NAPAP quantities being generally higher. Differences in Version 5.0 of the inventory are likely to be less. Emissions data in NAPAP Version 3.0 show reasonable agreement with emissions in Trends and Work Group 3B. Some comparisons may improve when Version 5.0 of the inventory is created.

Version 5.0 of the NAPAP emissions inventory represents the best detailed 1980 emissions inventory presently available.

Table 1

National Summary of Emissions
from all Versions of the 1980 NAPAP Inventory
(10^3 tons/year)

Pollutant	Emissions Inventory Version 1.0	2.0	3.0	4.0
SO_x	27,916	27,784	27,111	27,222
NO_x	22,905	23,198	23,667	23,833
VOC	22,582	22,520	23,269	23,307
SO_4	--	--	--	947
NH_3	--	--	--	4,250

Table 2

NAPAP Emissions by Source Category
(10^6 tons/year)

	SO_x	NO_x	VOC
Electric Utilities	17.3	8.1	0.1
Industrial Combustion	3.7	4.5	1.0
Residential/Commercial Combustion	0.9	0.7	0.1
Non-ferrous Smelters	1.2	Neg	Neg
Other Industrial Processes	3.0	1.0	4.5
Transportation	0.9	9.1	8.0
Miscellaneous	0.1	0.3	9.6
Total	27.1	23.7	23.3

Table 3

NAPAP Emissions by Region[a]
(10^6 tons/year)

	SO_x	NO_x	VOC
EPA Region 1	0.7	0.6	1.2
EPA Region 2	1.2	1.2	1.8
EPA Region 3	3.8	2.3	2.1
EPA Region 4	6.5	4.1	4.0
EPA Region 5	7.9	4.8	4.5
EPA Region 6	2.3	5.3	4.0
EPA Region 7	2.0	1.8	1.4
EPA Region 8	0.8	1.2	0.9
EPA Region 9	1.5	1.7	2.4
EPA Region 10	0.4	0.7	1.0
31 Eastern States[b]	22.3	15.1	15.3
Nation[a]	27.1	23.7	23.3

[a] Includes Continental U.S. only.

[b] Includes tier of states from Minnesota south to Louisiana and all states east.

Table 4

NAPAP Emissions by Release Height Range
(percent of total)

Range (ft)	SO_x	NO_x	VOC
0-120	26	68	99
121-240	14	8	1
241-480	20	10	0
>480	40	14	0
Total (10^6 tons/year)	27.1	23.7	23.3

Table 5

National Summary of Sulfate and Ammonia Emissions by Source Category
(10^3 tons/year)

Category	Emissions	
	SO_4	NH_3
Point Source		
Electric Utility Boilers	448	561
Industrial Boilers	172	63
Commercial/Institutional Boilers	25	3
Industrial Processes	149	219
Total	794	846
Area Source		
Stationary Fuel Combustion	118	16
Mobile Fuel Combustion	35	0
Field Application of Manure	0	3,311
Anhydrous Ammonia Fertilizer Application	0	53
Beef Cattle Feedlots	0	24
Total	153	3,404
Grand Total	947	4,250

Table 6

Summary of Canadian Emissions by Source Category
(10^3 tons/year)

Source Category	Pollutant Emissions					
	SO_x	NO_x	VOC	SO_4	NH_3	Alkaline
Electric Utilities	844	235	5	18	--	15
Non-Ferrous Smelters	1,507	---	---	29	--	--
Other Industrial Processes	1,754	114	417	87	20	96
Industrial Combustion	213	34	<1	6	--	1
Area Source Stationary Combustion	577	313	52	43	2	3
Transportation	151	1,181	878	5	--	<1
Miscellaneous	65	259	2,231	1	<1	6,840
Total	5,111	2,136	3,583	189	22	6,955

DEVELOPMENT OF THE NAPAP UTILITY REFERENCE FILE

by

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DEVELOPMENT OF THE NAPAP UTILITY REFERENCE FILE

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E.H. Pechan & Associates, Inc.

ABSTRACT

Emissions from electric utilities account for a large share of total acid deposition precursor emissions. The importance of the utility industry has been recognized by NAPAP through many of the activities of NAPAP's Task Group B (Man-Made Sources), which has sponsored work in both developing estimates of current emissions from the utility industry and projecting future emissions. Different components of NAPAP have overlapping but not identical data needs; the purpose of this project was to develop a single data base that would meet the utility-related data needs of all NAPAP task groups. Because existing data files were inadequate for this purpose, Task Group B requested that E.H. Pechan & Associates, Inc. (Pechan) develop a single, comprehensive utility-related data file. This paper documents the resulting data system, designated the NAPAP Utility Reference File (NURF).

DEVELOPMENT OF THE NAPAP UTILITY REFERENCE FILE

INTRODUCTION

Different components of NAPAP have overlapping but not identical data needs; the purpose of this project was to develop a single data base that would meet the utility-related data needs of all NAPAP task groups. Existing data files developed under Task Group B sponsorship were inadequate for this purpose. For example, Version 4.0 of the 1980 NAPAP emissions inventory (Toothman, 1984), between its point source and area source components, includes only anthropogenic sources of major acid deposition precursor pollutants for that year. As another example, the Unit Inventory (Pechan, 1983) was developed to meet data requirements of the Advanced Utility Simulation Model (AUSM); the Unit Inventory contains unit-specific information for only a subset of generating units, however. In response to the needs of NAPAP, Task Group B requested that E.H. Pechan & Associates, Inc. (Pechan) develop a single, comprehensive utility-related data file. This data base, designated the NAPAP Utility Reference File (NURF), will be used as an input to Task Group B utility emissions models.

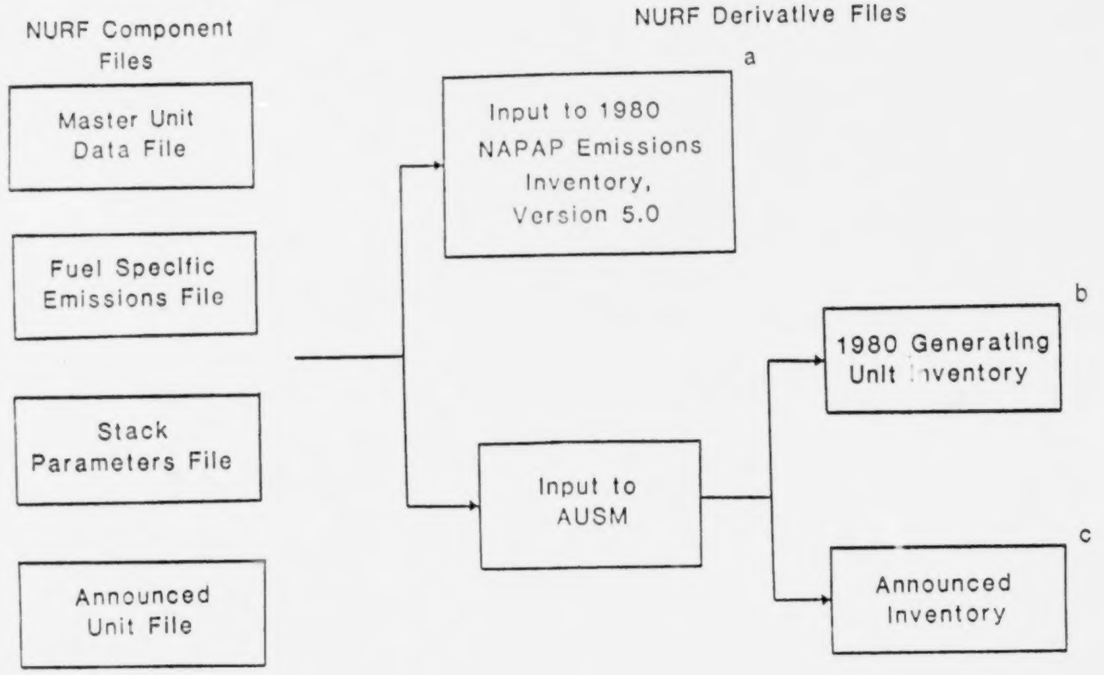
DISCUSSION

Figure 1 provides an overview of the NURF data system. NURF comprises four component files: the master unit data file (containing information on all electric utility generating units), the fuel specific emissions file (containing emissions information by fuel and unit), the stack parameters file (containing stack information on a unit-specific basis), and the announced unit file (containing unit-specific information on units coming on-line or converting to coal-firing after 1980). In addition, as shown in Figure 1, three files were derived from NURF and made available for distribution: the NAPAP emissions inventory updates file (containing corrections and addendums to Version 4.0 of the 1980 NAPAP emissions inventory), and information for AUSM, including a 1980 generating Unit Inventory (an updated version of the existing file containing unit-specific information on larger generating units and aggregated information on smaller units) and the announced inventory (containing information on units coming on-line or planned for conversion to coal after 1980).

NURF was developed using information from Version 4.0 of the 1980 NAPAP emissions inventory, the Unit Inventory, and new data; additional updates were obtained from a variety of sources. Table 1 lists major data inputs to NURF. In developing NURF, the greatest reliance was placed on data available from public sources, especially files available in machine-readable form. A variety of processing techniques were required to format

Figure 1

OVERVIEW OF NURF ELEMENTS


NURF Component Files
Master Unit Data File
Fuel Specific Emissions File
Stack Parameters File
Announced Unit File
NURF Derivative Files
Input to 1980 NAPAP Emissions Inventory, Version 5.0
a
Input to AUSM
1980 Generating Unit Inventory
b
Announced Inventory
c

[a] existing plants emitting SO_2, NO_x, or TSP

[b] existing plants aggregated

[c] planned plants (or conversions)

Note: The three NURF derivative files were made available for distribution.

Table 1

Major Data Inputs to NURF

Name	Type*	Level of Detail	Data Provided
EIA Form 759	A	Plant	Fuel consumption and generation
FPC Form 423	A	Plant	Cost and quality of fuels
FPC Form 67	A	Boiler/fuel feeder	Fuels consumed, control equipment, firing type, and bottom type
FGDIS	A	Boiler	SO_2 scrubber data
NAPAP Inventory	A	Boiler	Stack parameters
Generating Unit Reference File	A	Unit	Year online, year retired, capacity
EPA State Implementation Plan file	A	Unit	TSP, SO_2, and NO_x Emission Limits
NERC planned plants	M	Unit	New plants
NERC coal conversions	M	Unit	Plants converted or planning conversion to coal
DOE coal conversions	M	Unit	Plants converted or planning conversion to coal
Pechan nuclear plant status	M	Unit	Status of planned nuclear units
NO_x control update	M	Unit	NO_x control measures
ANL firing data	A	Unit	Supplemental data on firing types and bottom type

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*A=Automated, M=Manual

the available information from these sources into a single consistent data system.

Other data elements (e.g., heat rate and capacity factor) were simply not available from standard sources and, therefore, had to be calculated from data that were available. For those cases in which the calculated values were unreasonable, judgements were made as to which of the input values were the most accurate; other data were adjusted on a unit-specific basis so that all resultant values were reasonable.

Because of the wide range of types of data processing required to create NURF, a combination of programming languages and methodologies was used in its development. Initial data reduction of the larger data files was performed by a series of programs written in Programming Language/I (PL/I). These larger files were reduced to one record per unit (or plant if a plant-level file). The reduced files were converted to a consistent format using the Statistical Analysis System (SAS). These SAS files were then combined and manipulated using a series of SAS programs. Examples of the processing required include calculation of default state average fuel quality data and assignment of plant technology codes. The result of these manipulations was a final NURF in SAS format. The final processing steps converted the NURF data in their component file form into the set of derivative files which were discussed earlier.

Development of NURF was a multi-step process. The first step was to identify the universe of units to be included. Because the coverage of the major contributing data files differs, it was necessary to compare several data files on a plant-specific basis in order to develop the best possible universe of facilities. Conflicts between these files occurred in many cases. These had to be resolved on a case-by-case basis. In cases where a conflict occurred, if two or more files contained consistent information, those data were chosen. Otherwise, preference was given to the U.S. Department of Energy's (DOE) 1980 Form 67 data (U.S. DOE, 1980b) and the Generating Unit Reference File (GURF -- U.S. DOE, 1981b), both of which contain the most recently updated information for 1980.

Operating characteristics such as total generation and total fuels consumed for generation were obtained from DOE's Form 4 (U.S. DOE, 1980a). Form 4 is a monthly data base; for use in NURF, however, the monthly data for 1980 were aggregated into annual totals. Form 4 data are reported by plant and prime mover. For example, a plant having both steam and combustion turbine units would have fuel use and generation data provided for each of these two prime movers.

Unit-specific operational data for 1980 were estimated by applying 1980 unit shares computed from 1980 Form 67 data on fuel use and generation to the corresponding data elements from the 1980 plant and prime mover totals from Form 4. This method was adopted to correct errors associated with the use of incorrectly reported units on Form 67 submissions. Because the Form 67 data were used only to compute shares of validated totals, unit errors in Form 67 data were not carried through to NURF.

Fuel quality data were obtained from the Federal Power Commission (FPC) Form 423 (U.S. DOE, 1981a). For units with no reported Form 423 information, state average data obtained from Form 423 were used.

CONCLUSIONS

NURF is a central repository of data for utility operations and emissions, making analysis of the electric utility sector convenient. The largest sources of generated power and sulfur dioxide emissions have been thoroughly covered. In addition, data on all large units (including fossil-fired, nonfossil-fired, existing, and planned) are provided in a comprehensive manner.

The major user files derived from NURF are inputs to Version 5.0 of the 1980 NAPAP emissions inventory and the 1980 generating Unit Inventory, an input to AUSM. Due to data coverage and desired data base size considerations, both of these derivative files have been segmented into components comprising large and small generating units. The size cutoff utilized in the Unit Inventory is more restrictive than that used to update Version 4.0 of the 1980 NAPAP emissions inventory because the Unit Inventory is used as the basis for projection of emissions and fuel use in AUSM; detailed projections of the activity of smaller units cannot reasonably be made. For example, the 1980 generating Unit Inventory includes unit-specific data on only 30 percent of all fossil-fired units, while Version 5.0 of the 1980 NAPAP emissions inventory contains detailed data on 62 percent of all fossil-fired units; because the units represented in detail in these data files are the larger units, these files have more extensive coverage of generating capacity, generation, and emissions. For fossil-fired generating capacity, 92 percent of the total is represented in unit-specific data in the 1980 generating Unit Inventory and 96 percent of the total is included in Version 5.0 of the 1980 NAPAP emissions inventory file. Generation and emissions coverage is almost complete for Version 5.0 of the 1980 NAPAP emissions inventory file and is extremely high (more than 98 percent) for the 1980 generating Unit Inventory. Note that the Unit Inventory also includes nonfossil units, which are not included in the NAPAP inventory.

Table 2 provides some summary statistics on NURF's coverage of generation capacity, generation, and emissions, by fuel and plant type. Fossil-fired units account for more than 75 percent of capacity and generation, and 100 percent of emissions. Table 2 also shows that the proportion of coal-fired generation is larger than the proportion of coal-fired capacity; the opposite is true for oil-fired units. As expected, coal-fired units

Table 2

Summary Statistics by Plant Type

Plant Type	Capacity (MW)	% of Total	Generation (TWh)	% of Total	SO_2 (10^3 tons)	% of Total	NO_x (10^3 tons)	% of Total	TSP (10^3 tons)	% of Total
Coal steam	242,700	42	1,171,000	51	16,070	91	5,200	79	570	88
Oil steam	67,600	11	217,000	10	1,320	8	400	6	60	9
Gas steam	93,500	16	338,000	15	120	1	750	12	20	3
Oil nonsteam	29,300	5	7,000	0	10	0	40	1	3	0
Gas nonsteam	24,400	4	21,000	1	0	0	100	2	2	0
TOTAL FOSSIL	457,500	78	1,754,000	77	17,520	100	6,490	100	655	100
Nuclear	48,900	8	233,000	10	0	0	0	0	0	0
Hydroelectric	81,500	14	287,000	13	0	0	0	0	0	0
Other	900	0	5,000	0	0	0	0	0	0	0
TOTAL	588,800	100	2,279,000	100	17,520	100	6,490	100	655	100

Note: Percentages calculated from detailed data.

account for the majority of emissions.

NURF was compiled from every major publicly available data file. By matching and combining this information, the development of NURF gathered all relevant information into one data file. Discrepancies that existed in the past between data bases are resolved in NURF; the result is a cohesive and comprehensive electric utility operations and emissions data file. Because NURF is a composite of the best data available from a number of sources, it does not exactly match any other individual file. Aggregate measures of key variables are in excellent agreement with published statistics, however. NURF is important as an input to NAPAP's emission efforts, as well as an important utility operations data base in its own right.

REFERENCES

Pechan, 1983: E.H. Pechan & Associates, Inc., "Electric Utility Unit Inventory: Database Technical Documentation," prepared for University of Illinois at Urbana Champaign, May 1983.

Toothman, 1984: Toothman, Douglas A., John C. Yates, and Edward J. Sabo, "Status Report on the Development of the NAPAP Emission Inventory for the 1980 Base Year and Summary of Preliminary Data," EPA-600/7-84-091 (NTIS No. PB85-167930), December 1984.

U.S. DOE, 1980a: U.S. Department of Energy, "Monthly Power Plant Report," EIA-759 data file, 1980.

U.S. DOE, 1980b: U.S. Department of Energy, "Steam-Electric Plant Operation and Design Report," EIA-767 data file, 1980.

U.S. DOE, 1981a: U.S. Department of Energy, "Cost and Quality of Fuels for Electric Utility Plants," DOE/EIA-0075(80), May 1981.

U.S. DOE, 1981b: U.S. Department of Energy, "Inventory of Power Plants in the United States: December 1980," DOE/EIA-0348(83), 1984.

DEVELOPMENT OF AN EMISSIONS INVENTORY TO SUPPORT TESTING OF THE EULERIAN REGIONAL ACID DEPOSITION MODEL

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EPA Contract No. 68-02-3997
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ABSTRACT

One of the most extensive applications of the National Acid Precipitation Assessment Program (NAPAP) Emissions Inventory is to support development and testing of the Eulerian Regional Acid Deposition Model (RADM). To accomplish this, the 1980 NAPAP Emissions Inventory had to be further resolved temporally, spatially, and by component species. Annual emissions had to be allocated to hourly for a typical weekday, Saturday, and Sunday in each season; county-level area sources and minor point sources had to be spatially resolved to modeling grid cells; NO_x emissions had to be split into NO and NO_2; TSP emissions had to be assigned to alkalinity classes; and VOC emissions had to be apportioned into 30 photochemical reactivity classes. To create the NAPAP Eulerian Modeling Emissions Inventory, sets of temporal, spatial, and pollutant species allocation factors were developed and applied to the annual data. This paper presents a discussion of the processes, assumptions, and data sources used to generate the 1980 NAPAP Eulerian Modeling Emissions Inventory and presents results of that effort.

DEVELOPMENT OF AN EMISSIONS INVENTORY TO SUPPORT TESTING OF THE EULERIAN REGIONAL ACID DEPOSITION MODEL

INTRODUCTION

The 1980 NAPAP Emissions Inventory contains annual emissions from point and area sources, with area sources compiled on a county basis. To support testing and development of the Eulerian Regional Acid Deposition Model (RADM):

- annual emissions had to be further resolved to hourly values;
- county-level area source data had to be spatially allocated to grid cells;
- NO_x emissions had to be split into NO and NO_2;
- TSP emissions had to be assigned to alkalinity classes; and
- VOC emissions had to be assigned to photochemical reactivity classes.

To create Version 5.2 of the 1980 NAPAP Eulerian Modeling Emissions Inventory, sets of temporal, spatial, and pollutant species allocation factors were developed and applied to the annual data contained in the 1980 NAPAP Emissions Inventory. Development of a more efficient and flexible data handling system, the Flexible Regional Emissions Data System (FREDS), was also required. Development of the 1980 NAPAP Eulerian Modeling Emissions Inventory, the associated allocation factors, and the data handling system is discussed below.

DISCUSSION

Temporal Allocation

Temporal allocation of the NAPAP emissions data was accomplished by applying a set of seasonal, daily, and hourly fractions to the annual emissions totals to generate hourly emissions values for a typical weekday, Saturday, and Sunday in each season.

For area sources, temporal allocation factors were derived based on published activity statistics (e.g., heating degree days, gasoline sales, air traffic activity). Where such data were unavailable, patterns for similar studies were utilized or engineering judgement was employed.

For electric utilities in the Eastern United States, State and SCC-specific hourly temporal factors were developed based on hourly power plant fuel use data previously collected for use in EPRI's SURE Program. The SURE data were also used to derive SCC-specific patterns for the rest of the United States. For other point sources, point-specific patterns were derived based on operating rate information contained in the annual inventory records.

Since the NAPAP study area spans several time zones, temporal allocation factors were adjusted to represent Greenwich Mean Time. Adjustments were also made to reflect Daylight Savings Time during the appropriate seasons in applicable areas.

Spatial Allocation

The NAPAP modeling grid system (Version 5.2) is comprised of 63,000 grid cells (210 rows, 300 columns) dimensioned 1/6° latitude by 1/4° longitude (approximately 20 x 20 km). Point source records in the NAPAP Emissions Inventory contain locational information making their assignment to grid cells straightforward. Area source emissions are allocated from counties to grid cells using spatial allocation factors, fractional multipliers that assign a portion of each county's emissions to a particular grid cell. Generally, since the subcounty distribution of emissions is unknown, emissions are apportioned on the basis of the known distribution of some surrogate indicator.

Spatial allocation factors were derived for population, housing, total land area, 10 land use classifications, and 1 composite land use category. Population and housing counts from the 1980 Census were obtained by enumeration district. Spatial allocation factors were created for those two surrogates by assigning populations and housing units to grid cells based on the location of the centroid of each enumeration district.

Land use data, derived from Landsat mosaic images, were obtained and contain 10 land cover/use percentages for each NAPAP grid cell. These land use percentages were utilized with grid/county land area relationships to derive spatial allocation factors. The final step in spatially allocating county-level emissions to grid cells was to assign the most appropriate of the 14 possible surrogates to each of the 88 area source categories.

Species Allocation

To be used in the RADM, three of the pollutants in the NAPAP Emissions Inventory, NO_x, TSP, and VOC, had to be further refined. NO_x emissions had to be split into NO and NO_2, TSP emissions assigned to alkalinity classes, and VOC emissions broken down by photochemical reactivity class.

Profiles of NO_x emissions, referenced by Source Classification Code (SCC), were developed using patterns previously derived for the SURE and Northeast Corridor Regional Modeling Project (NECRMP) inventories. Since the modelers desire the flexibility to test a number of different chemistries, a more complex approach was necessitated for VOC emissions. Using primarily published information, a set of species profiles were developed. Each profile provides a breakdown of a category's VOC emissions into a typical set of component species. By distributing emissions to individual species, the profiles become independent of any reactivity scheme, providing the flexibility desired by the modeling community. To "speciate" emissions, two additional files were employed. First, the SCC Index File matches each SCC with the most appropriate profile. Next, the Class Assignments File assigns each of the 160 species contained in the profiles to one of the 30 photochemical classes specified by the modelers. Alkaline dust emission

Spatial allocation factors were derived for population, housing, total land area, 10 land use classifications, and 1 composite land use category. Population and housing counts from the 1980 Census were obtained by enumeration district. Spatial allocation factors were created for those two surrogates by assigning populations and housing units to grid cells based on the location of the centroid of each enumeration district.

Land use data, derived from Landsat mosaic images, were obtained and contain 10 land cover/use percentages for each NAPAP grid cell. These land use percentages were utilized with grid/county land area relationships to derive spatial allocation factors. The final step in spatially allocating county-level emissions to grid cells was to assign the most appropriate of the 14 possible surrogates to each of the 88 area source categories.

Species Allocation

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Eulerian Modeling Files

In addition to development of allocation factors to address the above requirements, a data handling system capable of creating subsequent versions of the NAPAP Emissions Inventory has been developed. The Flexible Regional Emissions Data System (FREDS), consisting of six primary independent subsystems written in the command language of the Statistical Analysis System (SAS), is being used to preprocess EIS formatted emissions data for use in regional scale atmospheric simulation modeling. The function of each module and its relationship to the system as a whole are presented in Figure 2.

In addition to preprocessing annual emissions data in Emissions Inventory System (EIS) Master File format and allocating these emissions by time, pollutant species, and location, FREDS calculates emissions uncertainty at any given level of temporal, species, and spatial allocation. For example, percent error values can be assigned to annual emissions which have not been allocated in space, time, or by pollutant species. On the other hand, the percent error values can be assigned for SCC level emissions which have been fully allocated.

RESULTS

Hourly emissions of SO_x, NO_x, VOC, and NH_3 for a typical summer weekday, nationwide, are shown in Figure 3. Figure 4 shows the distribution of VOC emissions by reactivity class as a function of time for a typical summer weekday. Spatial distribution of SO_x emissions for 1600 Greenwich Mean Time of a typical summer weekday is depicted in Figure 5. (Note that, for ease of interpretation, each grid cell depicted in Figure 5 actually represents 24 of the NAPAP grid cells.)

CONCLUSIONS

An emissions data base with the spatial, temporal, and species resolution necessary to support development and testing of the Eulerian Regional Acid Deposition Model (RADM) has been developed. A corresponding data handling system, which addresses the extensive requirements of the RADM, has also been developed and provides substantial improvements in efficiency over the predecessor system.

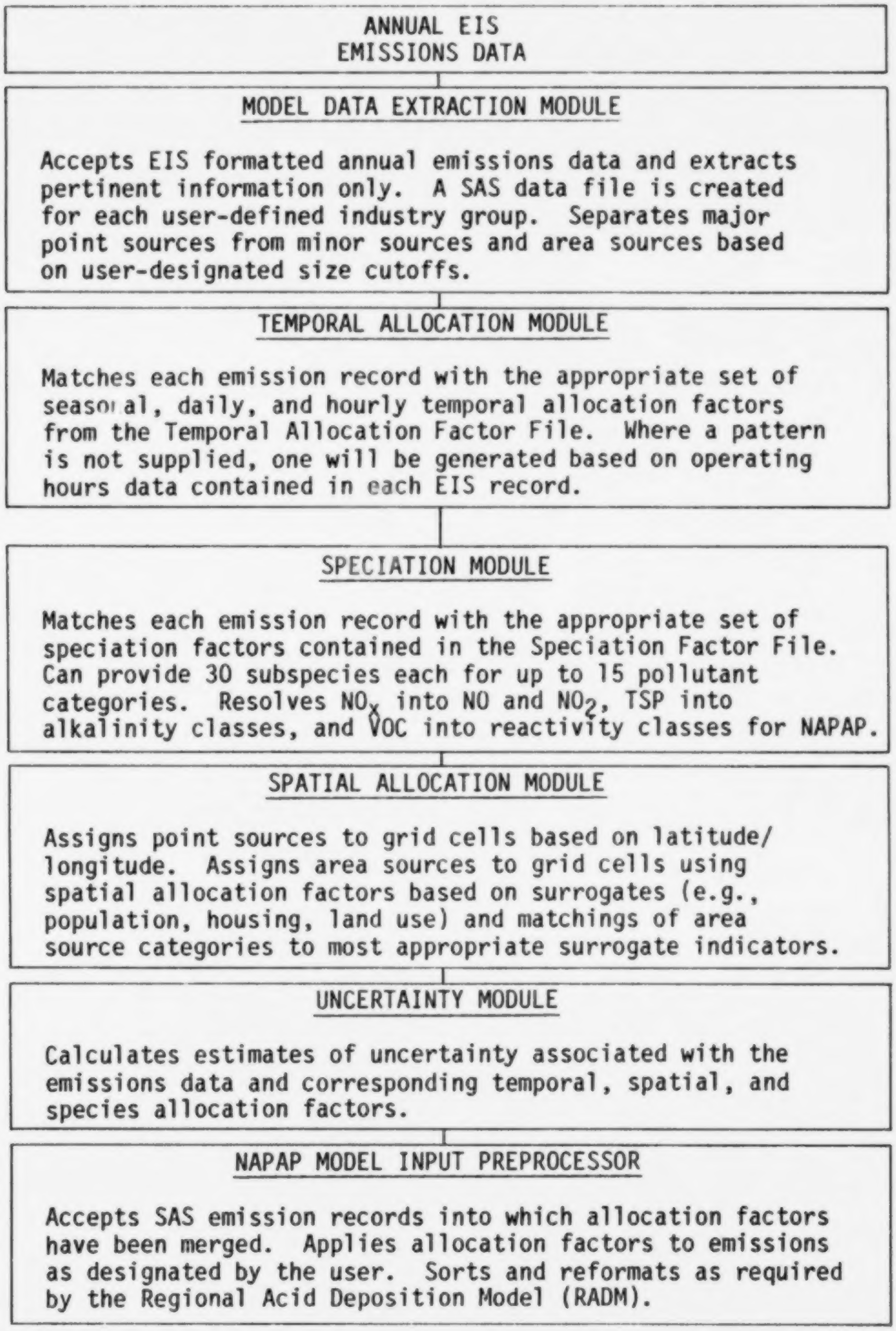
TEMPORAL RESOLUTION: Hourly emissions values for a typical weekday, Saturday, and Sunday for all four seasons

GEOGRAPHIC DOMAIN/RESOLUTION: 48 States and Canada; separate major point source file with minor point sources and area sources assigned to 63,000 20 x 20 km grid cells

POLLUTANTS

- SO_2
- SO_4
- TSP:
  - Ca
  - Mg
  - K
  - Na
- Pb
- CO
- HCl
- HF
- NO
- NO_2
- NH_3
- VOC:
  - Methane
  - Ethane
  - Ethylene
  - Propane
  - Propylene
  - N-butane
  - 1,2-butene
  - Isobutane
- VOC (cont.)
  - Isobutene
  - Trans-2 butene
  - Pentane
  - Isopentane
  - 2,3-dimethylbutane
  - Other alkenes
  - Other alkanes
  - Formic acid
  - Acetic acid
  - Other organic acids
  - Formaldehyde
  - Acetaldehyde
  - Propionaldehyde
  - Acetone
  - Other ketones
  - Other aldehydes
  - Xylene
  - Benzene
  - Toluene
  - Ethylbenzene
  - Other aromatics

Figure 1. Contents of the 1980 NAPAP Eulerian Modeling Emissions Inventory.



ANNUAL EIS
EMISSIONS DATA

MODEL DATA EXTRACTION MODULE

Accepts EIS formatted annual emissions data and extracts pertinent information only. A SAS data file is created for each user-defined industry group. Separates major point sources from minor sources and area sources based on user-designated size cutoffs.

TEMPORAL ALLOCATION MODULE

Matches each emission record with the appropriate set of seasonal, daily, and hourly temporal allocation factors from the Temporal Allocation Factor File. Where a pattern is not supplied, one will be generated based on operating hours data contained in each EIS record.

SPECIATION MODULE

Matches each emission record with the appropriate set of speciation factors contained in the Speciation Factor File. Can provide 30 subspecies each for up to 15 pollutant categories. Resolves NO_x into NO and NO_2, TSP into alkalinity classes, and VOC into reactivity classes for NAPAP.

SPATIAL ALLOCATION MODULE

Assigns point sources to grid cells based on latitude/longitude. Assigns area sources to grid cells using spatial allocation factors based on surrogates (e.g., population, housing, land use) and matchings of area source categories to most appropriate surrogate indicators.

UNCERTAINTY MODULE

Calculates estimates of uncertainty associated with the emissions data and corresponding temporal, spatial, and species allocation factors.

NAPAP MODEL INPUT PREPROCESSOR

Accepts SAS emission records into which allocation factors have been merged. Applies allocation factors to emissions as designated by the user. Sorts and reformats as required by the Regional Acid Deposition Model (RADM).

Figure 2. Major components of the Flexible Regional Emissions Data System.

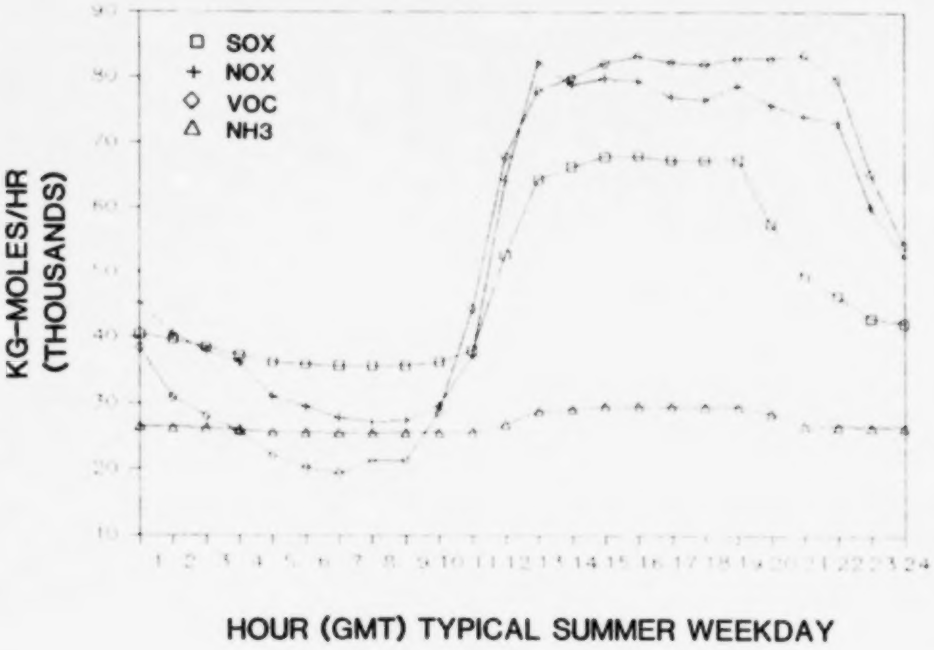


Figure 3. Diurnal emissions (1980) for a typical summer weekday, contiguous United States.

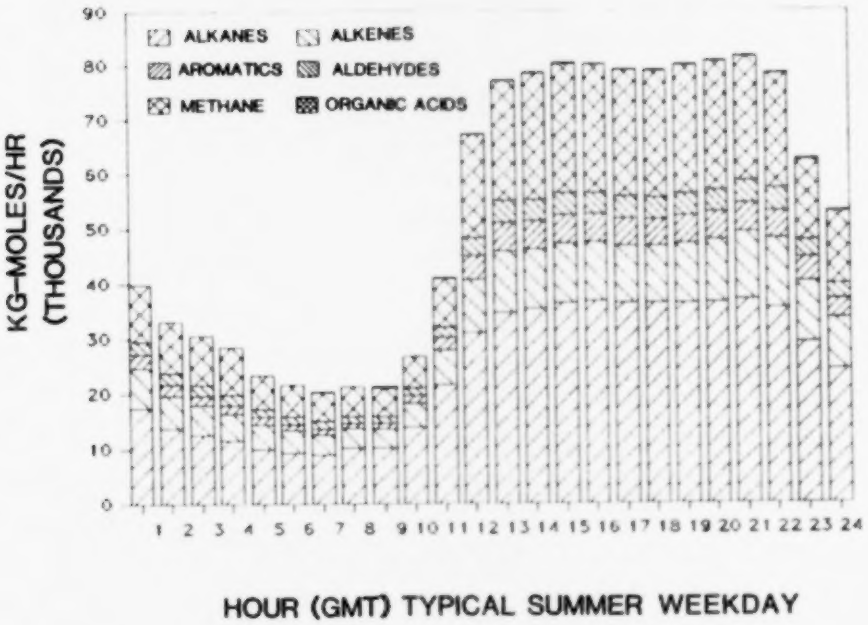


Figure 4. Diurnal VOC distribution by reactivity class for a typical summer weekday, contiguous United States.

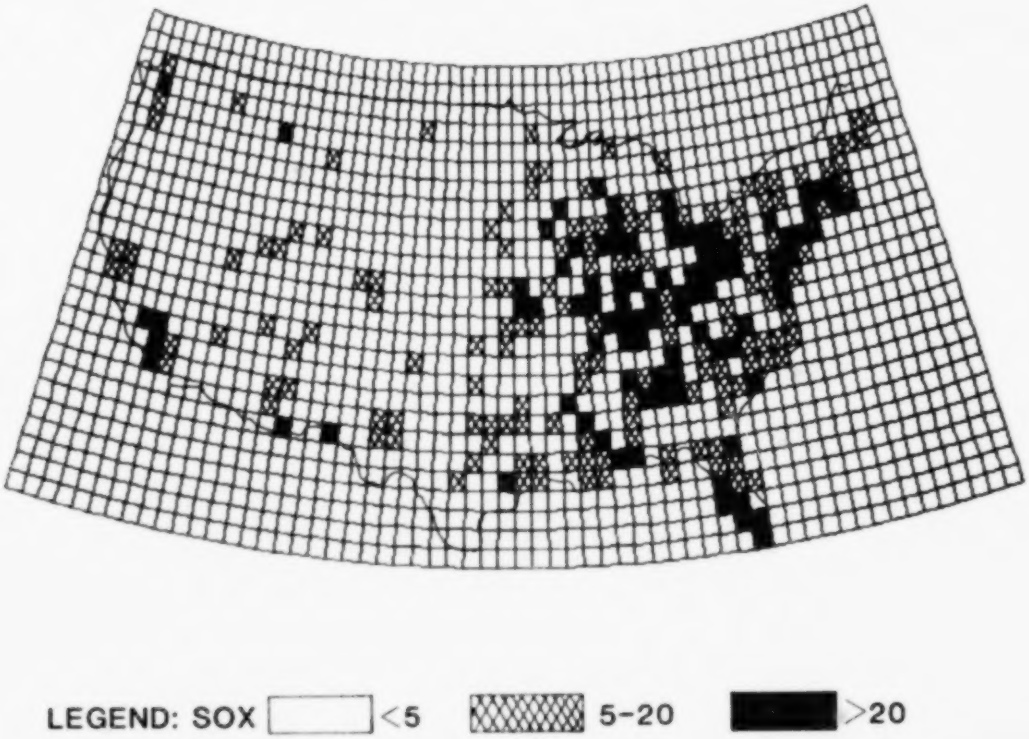

LEGEND: SOX
<5
5-20
>20

Figure 5. SO_x emission density (g-moles/hr/km^2), 1600 GMT for a typical summer weekday.

UNCERTAINTY ANALYSIS OF NAPAP EMISSIONS INVENTORY

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UNCERTAINTY ANALYSIS OF NAPAP EMISSIONS INVENTORY

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ABSTRACT

A major goal of Task Group B is the development and maintenance of detailed inventories of anthropogenic emissions to support acid deposition research. Estimates of the uncertainties of the emissions values are an integral part of any inventory developed. Detailed quantification of these uncertainties in the scale required by NAPAP researchers has never been attempted before. Methods used to estimate emissions were studied, and statistical methodologies were derived which will provide the best estimate of the uncertainties associated with these emissions values. These methodologies do not rely on any assumptions as to the statistical properties of the data needed to quantify the emissions uncertainties. Auxiliary data needed to estimate the uncertainties have been identified. A basic framework computer software has been developed to allow the calculation of the uncertainties associated with the emissions values in the NAPAP inventory. A workshop was held during which expert opinion was solicited, and the Delphi method was applied to arrive at quantitative estimates for the auxiliary data needed to calculate the uncertainties of emissions values for the NAPAP 1980 base year. Statistical formulations have been developed to incorporate these data into the methodologies selected to estimate the uncertainties associated with the emissions values. Development of the computerized data base and calculation of the emissions uncertainties are proceeding. Methodologies are being developed for the use of expert opinion to generate the auxiliary data needed to calculate emissions uncertainties for the NAPAP 1985 base year. Calculation of the 1985 base year uncertainties is expected to follow the 1985 data base development.

INTRODUCTION

A major goal of Task Group B is the development and maintenance of detailed inventories of anthropogenic emissions to support both assessment of the effects of acid deposition and the development, evaluation, and use of long range transport and transformation models for acid deposition research. Estimates of the uncertainties of the emissions values are an integral part of any inventory developed. The objectives of this project are:

1. To develop a methodologies framework to assess the uncertainties associated with the emissions data as presented in the NAPAP emissions inventory.

2. To develop and implement a prototype software package to calculate the uncertainties associated with the emissions values of the NAPAP emissions inventory.

3. To develop the data base needed to calculate the uncertainties associated with the values for the 1980 base year emissions.

4. To develop the data base needed to calculate the uncertainties associated with the emissions values for the 1985 base year.

DISCUSSION

The NAPAP emissions inventory was based on the National Emissions Data System (NEDS) currently operated by the Office of Air Quality Planning and Standards (OAQPS) of EPA. NEDS provided the basic data from which all other levels of aggregation or disaggregation will be calculated. The basic NEDS data are statistical averaged parameters which allow the calculation of yearly emissions of the five criteria pollutants (particulates, SO_x, NO_x, hydrocarbons, and CO) on an individual source/process basis for point sources and on a county level for area sources. Current plans call for the application of spatial, temporal, and species disaggregation algorithms which will be based on disaggregation factors (or modifications thereof) developed for the Northeast Corridor Regional Modeling Project (NECRMP). Higher levels of aggregation will be calculated as sums of the basic NAPAP data.

For each individual point source/process, yearly emissions are estimated using one of the following methods:

1. Stack tests or other emission measurements

2. Material balance

3. Calculated using federal (AP-42) emission factors, where emission factors are the rate at which a pollutant is released as a result of some activity

4. Guess

5. Calculated using special emission factors.

Emissions estimated using Methods 3 and 5 are calculated using the equation:

$$EM = \sum_{i=1}^{P} (EF_i)(A_i)(F_i) \quad (C) \tag{1}$$

where

EM = total emissions from a point source of a given pollutant

P = number of processes contributing to emissions from point source

EF_i = emission factor for process i

A_i = ash or sulfur content of fuel used by process i (if appropriate)

F_i = material or fuel throughput for process i

C = (1-control efficiency of control equipment) for a given pollutant

Yearly emissions for area sources are calculated by geographic county using the equation:

$$EM_c = \sum_{a=1}^{A} EMa = \sum_{a=1}^{A} (EF_a)(A_a)(AC_a)(AD_a) \tag{2}$$

where

EM_c = area source emissions for county c

A = number of activities contributing to emissions in county c

EM_a = area source emissions from activity a for county c

EF_a = area source emission factor for activity a (this parameter can also be county specific)

A_a = ash or sulfur content of fuel used in activity a (if appropriate)

AC_a = appropriate measurement of level of activity a in county c

AD_a = adjustment factor for area source category

Several previous projects have addressed the problem of assigning, in statistical terms, quantitative values to the errors in emissions data. Final reports from the following projects were reviewed.

1. Weighted Sensitivity Analysis of Emissions Data, project conducted by IBM Corporation under contract to the Office of Air Quality Planning and Standards (OAQPS), EPA, July 1973.

2. Source Inventory and Emission Factor Analysis (SIEFA) project conducted by PEDCo - Environmental Specialists, Inc., under contract to OAQPS, September 1974.

3. Emissions Inventory for the SURE Region, project conducted by GCA Corporation under contract to the Electric Power Research Institute (EPRI), April 1981.

4. Emissions, Costs and Engineering Assessment, Work Group 3B, US-Canada Memorandum of Intent (MOI) on Transboundary Air Pollution, June 1982.

5. Preliminary Evaluation of Acidic Deposition Assessment Uncertainties, project conducted by Argonne National Laboratory (ANL) under contract to the U.S. Department of Energy, November 1982 (preliminary report).

All these projects have based their calculations on the statistical formulas for error propagation as derived for "small" values of the errors; i.e., the Taylor series expansion included only the first derivatives of the function. Some of the error values used in subsequent calculations have been as high as 80-90% of the mean.

In calculating the mean squared error of emissions estimated such as those above, we take the stance that the component parts of each expression are themselves estimated, and that these component estimates have known (or estimable) expected values and variances. Further, the estimation errors of the components are independent of one another. The mean squared error of the emissions estimate then follows from simple formulas giving variances of algebraic combinations of random variables; e.g., for N independent random variables $U_i, \ldots, U_N$:

$$V(\Sigma U_i) = \Sigma V(U_i) \tag{3}$$

$$V(\pi U_i) = \pi \; E^2(U_i) + V(U_i) \quad - \pi E^2 \tag{4}$$

Since the errors of estimation are presumably independent across geographic space, the mean squared error of the estimate of an aggregate is simply the sum of the mean squared errors of its components.

The second objective of this project addresses the design and implementation of the basic framework of computer software needed to calculate uncertainties associated with yearly emissions values for both point and area sources. The conceptual design is independent of the software system used to support the NAPAP inventory; the design and implementation of the software modules will allow portability between computer systems and will be as independent as possible of the current NAPAP inventory software system. Outputs generated by this software include values for the uncertainties of emissions at the point, plant, county, state, or national level for point sources and at the county, state, or national level for area sources.

Uncertainty values associated with each parameter used in calculating emission values are expected to be applicable to whole "classes" of these parameters rather than just to individual sources. To avoid needless repetition of data (with the associated increase in storage costs and the ever present danger of errors at update times), a file with uncertainty "profiles" was designed. When the data are developed, this file will contain all the variance information needed to calculate the uncertainty value associated with the yearly emissions of a point source or an area source category. Each uncertainty profile will be identified by a unique code; at retrieval time the appropriate uncertainty profiles can be referenced to allow the calculation of uncertainty values associated with the reported emissions. Figure 1 presents a schematic of this design.

Given the wide range of elements requiring uncertainty estimation for eventual use in estimating the uncertainty of the emissions values in the 1980 NAPAP Emissions Inventory, the use of a panel of experts was considered as an approach to begin the task of building the data base needed to perform these calculations. As the initial activity of Objective 3 for this project, an uncertainty workshop concept was formulated to convene a panel with expertise in many of the elements included in an emissions inventory uncertainty analysis. The 1980 NAPAP Emission Inventory Uncertainty Workshop was held at the EPA Air and Energy Engineering Research Laboratory, Research Triangle Park, North Carolina, on May 7, 1985. The panel was queried on individual uncertainty estimates for the elements discussed above which are part of the emissions estimation process. The Delphi technique was applied to elicit the experts' opinions in a systematic manner leading to useful results. This technique involves iterative questionnaires administered to individual experts; feedback of results accompanies each iteration of the questionnaire. This process continued until a convergence of opinion was reached. The end product is the consensus of experts, including their commentary, on each of the questionnaire items.

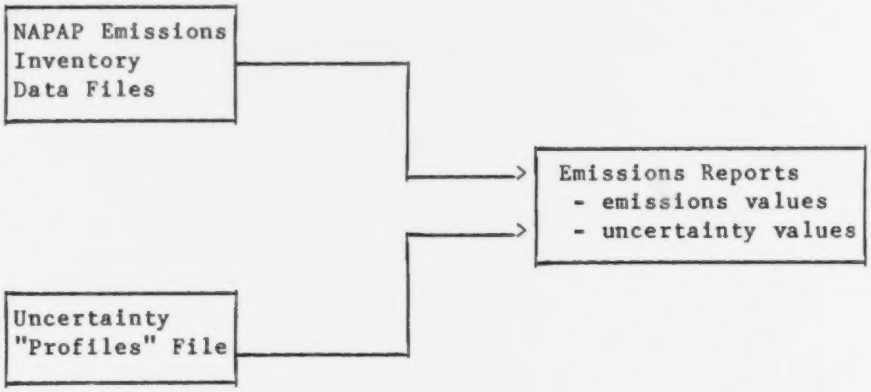
NAPAP Emissions Inventory Data Files
Uncertainty "Profiles" File
Emissions Reports
- emissions values
- uncertainty values

Figure 1
Software to Calculate Uncertainty Values

For each uncertainty value desired, the workshop participants were asked to estimate p in the statement: "90% of the data fall within p% of the mean." Table 1 summarizes the results of the workshop.

To allow the use of the values developed in the workshop with Equations (3) and (4) the concept of "fractional error" was developed. Suppose $\theta \sim N(\theta, \sigma^2)$ so that the quoted phrase above is equivalent to:

$$\Pr\left[(1-p)\,\theta \leq \hat{\theta} \leq (1+p)\,\theta\right] = 0.9$$

that is, $p = \frac{z\sigma}{\theta}$ where, for the normal distribution, $z = 1.6448$. We call p the "fractional error associated with $\hat{\theta}$ as an estimate of θ."

Fractional errors may be developed for algebraic combinations of independent random variables as follows:

$$P_x^2 \sum_{i=1}^{N} E(U_i)^2 = \sum_{i=1}^{N} P_{U_i}^2 \, E(U_i)^2 \qquad \text{for } X = \sum_{i=1}^{N} U_i \qquad (5)$$

$$P_x^2 = z^2 \prod_{i=1}^{N} (1 + k^2 P_{U_i}^2) - 1 \qquad \text{for } X = \prod_{i=1}^{N} U_i \qquad (6)$$

(Note that the relation between p and the variance depends on the normal assumption, and also that the product of normal random variables is not normal.)

Table 2 presents the distribution of confidence rating values of the point source emission factors by pollutant. Uncertainty profiles were developed based on the Delphi results, and a subset of the NAPAP 1980 base year Version 3 major point source file was used to perform initial calculations. The subset chosen included data for the state of Delaware and for the District of Columbia. Preliminary results indicate that, for annual point source emissions, calculated uncertainties are in the range of 20-100% for individual point sources and in the range 5-14% at the state level. Uncertainty profiles for annual emissions from area sources are currently being developed; uncertainty calculations will follow.

The use of expert teams will be extended in Objective 4 of this project. A draft plan for the development of the uncertainty values for the 1985 data base has been prepared. A finer breakdown of source categories and an increased number of pollutants are to be considered in the 1985 base year exercise. Four expert teams have been identified; these teams will address the utility, industrial combustion, industrial processes, and transportation sectors. Expert opinion will be obtained via individual interviews using the most appropriate formulation of probability encoding techniques. A classical statistical approach will be followed -- explicit specification of an underlying model followed by derivation of the problem

Table 1
Proposed Uncertainties for the NAPAP 1980 Base Year Inventory

Parameter	Uncertainty Range (%) SO_x	NO_x	VOC	Across all Pollutants
Fuel S content				10
Point source emission factors				
Method 1 (stack tests)	25	25	25	
2 (material balance)	10	25	50	
3 (AP-42 emission factors)				
confidence rating A	10	10	10	
B	25	25	25	
C	50	50	50	
D	75	75	75	
E	100	100	100	
5 (state emission factor)	25	50	50	
4,6,7,0 and blank	100	100	100	
Point source production throughput	15	15	15	
Control equipment efficiency	25	25	25	
Area source emission factors				
Mobile sources	*	50	100	
"Other" sources	25	25	100	
Area source activity level				
Mobile				25
Other				25
Point source temporal apportionment				
Seasonal profiles	10	10	10	
Daily profiles	25	25	25	
Hourly profiles	50	50	75	
Area sources temporal apportionment				
Seasonal profiles	10	10	10	
Daily profiles	25	25	25	
Hourly profiles	50	50	75	
Area sources spatial apportionment factors	25	25	25	
Chemical speciation factors				
NO/NO_2				25
VOC				100

*Apply same criteria as for point source emission factors.

Table 2

Emission Factor Confidence Level (from EIS Point Source Emission Factor File)

```
------------------------------------------------------------------------------
                                                                  CUMULATIVE
                                                         PERCENT  PERCENT
------------------------------------------------------------------------------
                                 Sulfur Oxides
   I
   -----------------------------------------+
 1 I                                    134 I            40.6      40.6
   -----------------------------------------+
   I
   ---------------------------+
 2 I                       90 I                          27.3      67.9
   ---------------------------+
   I
   ----------------------+
 3 I                  71 I                               21.5      89.4
   ----------------------+
   I
   ---------+
 4 I     27 I                                             8.2      97.6
   ---------+
   I
   --+
 5 I I 8                                                  2.4     100.0
   --+
   I
   I.........I.........I.........I.........I.........I
   0        10        20        30        40        50
   PERCENT

                                 Nitrogen Oxides
   I
   -----------------------------------------+
 1 I                                    104 I            34.7      34.7
   -----------------------------------------+
   I
   -----------------------------------+
 2 I                               88 I                  29.3      64.0
   -----------------------------------+
   I
   ---------------------------------+
 3 I                             84 I                    28.0      92.0
   ---------------------------------+
   I
   ---------+
 4 I     18 I                                             6.0      98.0
   ---------+
   I
   ---+
 5 I  I 6                                                 2.0     100.0
   ---+
   I
   I.........I.........I.........I.........I.........I
   0         8        16        24        32        40
   PERCENT
```

(Continued)

Table 2(continued)

```
                                                              CUMULATIVE
                                                     PERCENT  PERCENT
                   Volatile Organic Compounds

  I
  ---------------+
1 I          111 I                                      14.8    14.8
  ---------------+
  I
  -------------------------------------------------+
2 I                                            369 I    49.1    63.9
  -------------------------------------------------+
  I
  -----------------------+
3 I                  172 I                              22.9    86.8
  -----------------------+
  I
  -----------+
4 I       83 I                                          11.1    97.9
  -----------+
  I
  --+
5 I I 16                                                 2.1   100.0
  --+
  I
  I.........I.........I.........I.........I.........I
  0        10        20        30        40        50
  PERCENT
```

solution under the assumptions of the model. Towards this end, a documentation packet is being compiled to be sent to participants on expert teams prior to the interviews. All appropriate definitions, assumptions, and statistical questions will be addressed in the packet.

CONCLUSIONS

Strict estimation of the uncertainties associated with the emissions values as presented in the NAPAP emissions inventory requires the explicit specification of the sources of uncertainties in each method used to estimate these emissions and of the underlying statistical model and assumptions to be used in estimating the desired uncertainties. The development of the auxiliary data needed for the calculation of uncertainties associated with the 1980 base year emissions had to be carried out within restricted time and resource bounds. This exercise provided a basic framework within which to refine the techniques being developed to estimate the uncertainties of the 1985 base year emissions.

BIBLIOGRAPHY

Benkovitz, C.M., "Framework for Uncertainty Analysis of the NAPAP Emissions Inventory," EPA-600/7-85-036, NTIS-PB86-112570 (September 1985).

Ditto, F.H., L.T. Gutierrez, T.H. Lewis, and L.J. Rusbrook. Weighted Sensitivity Analysis of Emissions Data: Volumes I and II. EPA-450/3-74-022 a and b, NTIS-PB258413 and 258414, 1973.

Klimm, H.A. and R.J. Brennan. Emissions Inventory in the SURE Region. EPRI-EA-1913, 1981.

Knudson, D., M. Davis, J. Shannon, D. Sisterson, S. Viessman, M. Wisely, and R. Whitfield. Preliminary Evaluation of Acidic Deposition Assessment Uncertainties. ANL/EES-TM-XXX (Draft), 1982.

Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, "Compilation of Air Pollutant Emission Factors," third edition (plus supplements 8-15), AP-42, NTIS-PB275525 (1977).

Office of Air Quality Planning and Standards, Environmental Protection Agency. Source Inventory and Emission Factor Analysis. Volumes I and II, EPA-450/3-75-082 a and b, NTIS-PB247743 and 248829, 1974.

Rivers, M.E. and K.W. Riegel (Eds.). Emissions, Costs and Engineering Assessment Work Group 3B. United States-Canada Memorandum of Intent on Transboundary Air Pollution, 1982.

Toothman, D.A., Yates, J.C. and Sabo, E.J., "Status Report on the Development of the NAPAP Emission Inventory for the 1980 Base Year and Summary of Preliminary Data," EPA-600/7-84-091, NTIS-PB85-167930 (December 1984).

SESSION 2: DEVELOPMENT OF NAPAP EMISSION INVENTORIES FOR ANTHROPOGENIC SOURCES (continued)

Chairman: John Bosch, Chief
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APPLICATION OF THE NAPAP EMISSIONS INVENTORIES TO EULERIAN MODELING

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ABSTRACT

This paper presents the schedule for development and evaluation of Eulerian Acid Deposition Models as part of the National Acid Precipitation Assessment Program and discusses the critical need to reduce uncertainties in emissions inventories to provide more reliable modeling results for policy decision making. A strong recommendation is made to expand, improve, and standardize quality assurance procedures for emission inventory development and to incorporate knowledge gained through modeling sensitivity studies in ranking emissions inventory development activities.

INTRODUCTION

One of the primary goals of the National Acid Precipitation Assessment Program (NAPAP) is to develop a comprehensive Regional Acid Deposition Model (RADM) suitable for assessing source-receptor relationships. RADM also forms the scientific basis for development of a series of simpler "engineering models" useful for addressing policy and assessment questions. NAPAP has established the following schedule for development and evaluation of these Eulerian acid deposition models.

February 1986	Preliminary Evaluation of the First Generation Regional Acid Deposition Model (RADM)
October 1986	Testing and Preliminary Assessment of a sulfur deposition Engineering Model

October 1987	Testing and Evaluation of an Advanced Engineering Model Operational For Multiple Species
January 1988	Operational Second Generation RADM
March 1989	Evaluation of Second Generation RADM

NAPAP has planned two major assessments designed to provide decision makers with scientific information necessary for developing acid deposition control strategies. Both the Eulerian models and the emissions inventories will play an important role in the 1987 and 1989 assessments. The 1987 assessment will use an unevaluated set of models for preliminary regulatory analysis, whereas the 1989 assessment will use the evaluated models. Thus a major field study has been proposed for 1987 through early 1988 to collect data necessary for model evaluation. Knowledge of uncertainty both within the model and each associated input data set is required to assess confidence in model predictions. This paper will focus on uncertainty in the emissions data base as related to these Eulerian modeling efforts.

DISCUSSION

From a modeler's viewpoint, uncertainties in emission inventories can be categorized into two types: 1) uncertainties associated with errors or inadequacies in the data base, and 2) uncertainties associated with the representativeness of typical daily emissions patterns for any particular day being modeled. Both the emissions inventory developers and the modeling groups have begun to address the first type of uncertainty, but from different perspectives. The developers have been primarily concerned with improving and verifying those raw data elements which are used in estimating annual point and county level emissions. Those elements include emissions factors, fuel utilization and quality, production activity, and others. They have also been concerned with improving procedures and data necessary to provide the temporal, spatial, and species resolution required by the Eulerian models. Most of the quality control tests have been performed on these individual data elements and efforts are underway to associate a level of uncertainty with each of the individual elements comprising the final emissions estimates at point, county and grid level.

The modelers, on the other hand, primarily work with the emissions inventories in their most resolved form - hourly gridded emissions. Their quality control tests search mainly for individual grids that appear to be inconsistent with their surroundings, subregions of the modeling domain that appear unexpectedly high or low, VOC species ratios that seem inconsistent with ambient measurement data and unexpected temporal patterns within a subregion. Both the developers and the modelers have compared the NAPAP inventory with other inventories with somewhat different results because of the different frames of reference. Generally, the State totals for individual species favorably agree with other inventories, or discrepancies can be explained by differences in methodology. However, the modelers have found that even when State totals agree there are sometimes significant differences in the spatial or temporal patterns of the gridded emissions, and at times differences in the relative magnitudes of the reactive species. This contradiction is often caused by errors in the State emissions which have cancelled out in the State total. Thus it is important for the developers to begin to incorporate into their quality control procedures more analytical tests typically performed by the modelers. In addition, it is important for the modelers to have a better understanding of the quality control procedures and estimates of inherent variability of the individual data elements used in computing emissions.

Uncertainties in emissions introduced by the question of representativeness are crucial for some of the proposed model evaluation studies. The RADM will be examined in an episodic mode; thus, any major deviation of the estimated typical hourly emissions from actual emissions for the particular days being modeled has the potential to influence the model results. Additional burden is then placed on the inventory developers and modelers to ensure that there are no atypical operations in major sources during the modeling periods. For studies of the first generation RADM existing information on the major SO_2 point source emissions will be used to check the representativeness of the NAPAP emissions. Since the field study is still in the planning stages more options are available to reduce uncertainty in the emissions for the second generation RADM studies. Some of the choices include acquisition of data on any anomalous plant operation during the period of interest, installation and operation of continuous emissions monitors (CEM) on major sources, the collection of actual hourly fuel use or production data, improved activity data for area source emissions estimates, etc. All of these options involve significant additional cost beyond the production of a typical base year inventory. Therefore, there is a definite requirement to rank data needs to most effectively accommodate improvements in the base inventory as well as the specialized needs of episodic model studies. In response to this need, modeling studies using RADM are currently being performed to gain a better understanding of the sensitivity of the model to uncertainties in emissions. Attention is being given to spatial and temporal variability in all of the major chemical species, and speciation of VOC. These sensitivity studies should provide information useful to optimize the data collection activities and to reduce uncertainties in critical areas.

CONCLUSIONS

In order to confidently use emissions inventories in the development, evaluation, and application of large scale Eulerian models like RADM, it is essential to have an understanding of the quality of the input data bases. Standardized quality assurance and uncertainty estimation procedures must be established by the emission data base developers to provide emissions inventories that are reliable and directly useful for a variety of NAPAP applications. Current modeling sensitivity studies will provide feedback to effectively rank future emissions inventory improvement activities.

HISTORIC EMISSIONS OF SO_2 AND NO_x SINCE 1990

BY STACK HEIGHT RANGE AND BY SEASON

by

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ABSTRACT

Historic emissions of sulfur oxides and nitrogen oxides were estimated for each state of the conterminous United States. The emissions were estimated by individual source category on the state level from 1900 to 1980 for every fifth year and for 1978. The source categories included power plants, industrial boilers, industrial processes, commercial and residential heaters, natural gas pipelines, highway vehicles, off-highway diesel engines, and all other anthropogenic sources. These emissions were calculated from salient statistics indicative of fuel consumption or industrial output, estimations of average statewide fuel properties, and estimations of emission factors specific to each source category. The emission estimates were then aggregated to show the emission trends by state, region, and all states combined.

This paper summarizes the national historic emission trends by emission release (stack) height and by season. The paper outlines the approach taken and presents general results.

HISTORIC EMISSIONS OF SO_2 AND NO_x SINCE 1900

INTRODUCTION

Sulfur dioxide (SO_2) and nitrogen oxides (NO_x) are considered primary precursors of acidic precipitation. The anthropogenic emissions of these pollutants are suspected causes of many biological and chemical effects observed in recent years. Understanding the historic emission trends is important to understanding the development of acid-precipitation-related problems and causes of observed environmental effects.

Average emission rates for each study year were calculated for individual source categories for each state. The source categories are listed in Table 1 according to the fuel consumed. These categories represent all types of boilers, furnaces, engines, processes, and other anthropogenic emission sources. The basic steps involved in calculating state emissions have been described in detail by Gschwandtner[1] and by Gschwandtner et al.[2] The emissions by source category were totaled by year to provide an estimate of overall national emission trends.

Analysis by Release (Stack) Height

The percentage distribution of national emissions was then estimated for each source category according to four broad ranges of stack heights. In the case of electric utilities, individual power plant emission estimates and stack height data were used to determine the national distribution by height from 1950 to 1980. For earlier years and for other sources, a general trend in the percentage of emissions by stack height range was estimated based largely on engineering judgment. It was assumed that the percentage emissions of each category changed linearly over time. The percentages estimated for each year

TABLE 1. FUEL TYPES AND EMISSION SOURCE CATEGORIES

Bituminous Coal:	Electric Utilities Industrial Boilers and Space Heaters Commercial and Residential Uses Steam Railroads Coke Plants
Anthracite Coal:	All Uses
Residual Oil:	Electric Utilities Industrial Boilers and Space Heaters Commercial and Residential Uses Vessels
Distillate Oil:	Electric Utilities Industrial Boilers and Space Heaters Commercial and Residential Uses Railroads Vessels
Natural Gas:	Electric Utilities Industrial Boilers and Space Heaters Pipeline Compression Stations Commercial and Residential Uses
Wood:	Electric Utilities Industrial Boilers and Space Heaters Commercial Heating Residential Wood Stoves and Fireplaces
Gasoline and Diesel Fuel:	Highway Vehicles Off-Highway Vehicles Vessels
Other:	Wildfires Cement Plants Copper, Lead, and Zinc Smelters Miscellaneous Industrial Processes Miscellaneous Other Sources

were then multiplied by the total emissions of each category. These results provide a general indication of the national trend.

Analysis by Season

The seasonal emissions were also estimated for major source categories. These estimates were based on monthly fuel data available for only certain years, and an assumed percentage distribution of the emissions by season over time. The distribution for each category was weighted by the historic emission estimates to yield an average seasonal distribution for each study year for all categories combined.

DISCUSSION

Distribution by Release (Stack) Height

Figure 1 shows the overall national trend of SO_2 and NO_x emissions by stack height range. This analysis does not include stack exit velocities or atmospheric mixing heights, which are also important considerations. The analysis in this study suggests that more SO_2 emissions were released into the atmosphere from stacks taller than 240 ft* than from stacks shorter than this height since about 1945. By 1980, approximately 30 percent of the SO_2 emissions were emitted above 480 ft, for example, compared to only about 5 percent above this height in 1950. In 1950, six power plants reported a maximum stack height greater than 480 ft. Not only have the percentages increased, but total national SO_2 emissions also increased and peaked around 1970. The percentage of the total SO_2 emissions released below 120 ft has generally decreased over the study period. The distribution of NO_x emissions has historically remained constant, although on the national level the total emissions have steadily increased. Approximately 60 percent of the total NO_x emissions in 1980 were released from ground level sources; predominantly from transportation sources.

(*) 1 ft = 0.3048m

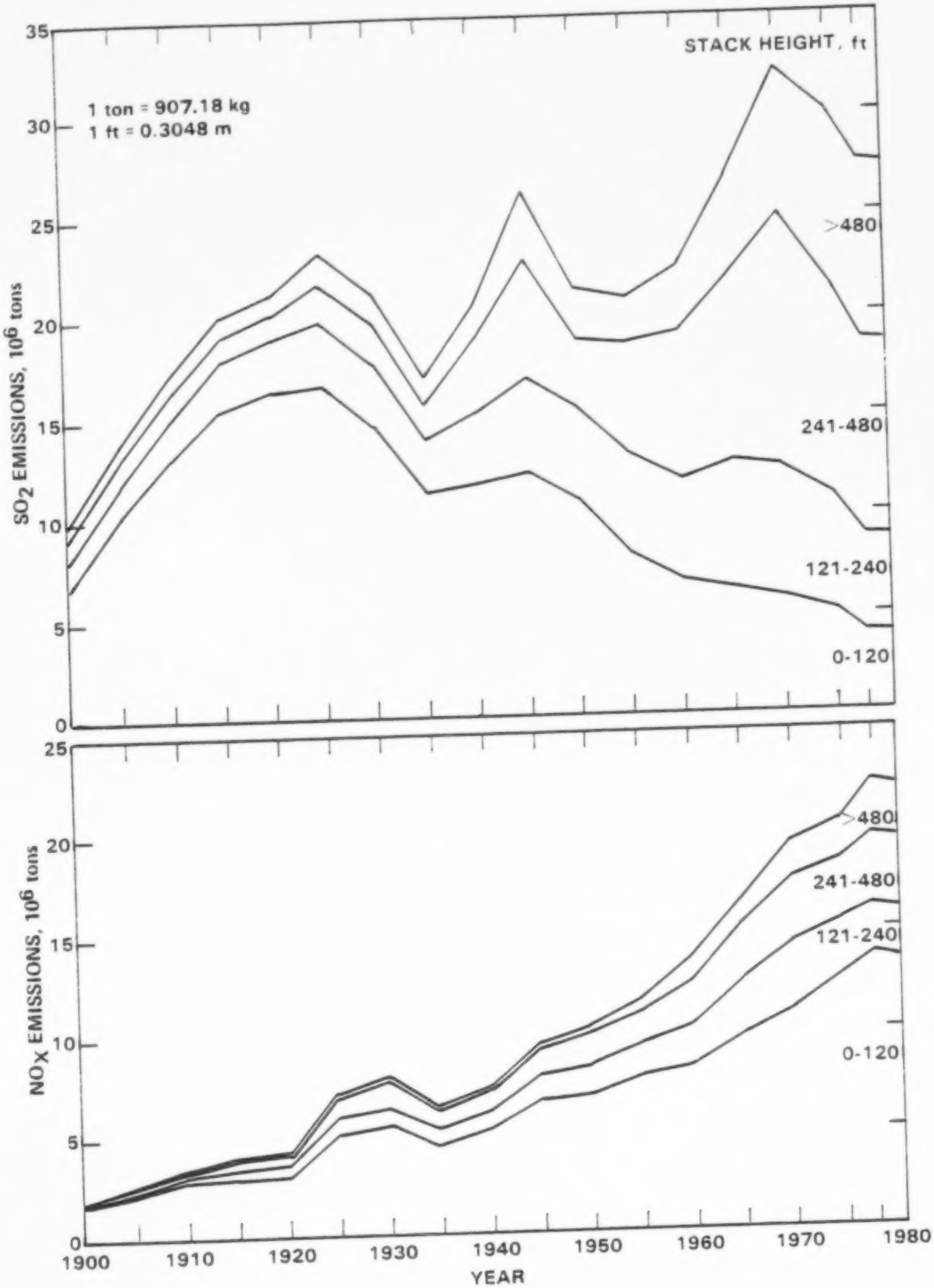

Figure 1. Total national SO$_2$ and NO$_X$ emissions by stack height ranges.

Analysis of the electric utility category alone suggests that, in the 1950's and 60's, most of the SO_2 and NO_x emissions from this category were released below 480 ft -- mostly between 240 and 480 ft. By 1980, about 50 percent of the total SO_2 emissions and 40 percent of the NO_x emissions from this source category were released above 480 ft as a result of the trend toward taller stacks. Since the emissions from electric utilities constitute a large portion of the total national emissions in recent years, as shown by Gschwandtner et al,[2] they have a significant effect on the overall distribution of emissions by release height.

Distribution by Season

Table 2 presents the estimated percentage distribution of emissions by season on the national level. The percentage of wintertime SO_2 emissions appears to have decreased from about 35 percent of the total annual emissions in 1900 to about 26 percent of the total in 1980. This gradual decrease is largely a result of switching from coal to fuel oil and gas for heating, and also an increase in summertime fuel combustion by power plants to meet the demands of refrigeration and air conditioning. The seasonal distribution is more pronounced on a regional level where climatic conditions deviate from the national average. NO_x emissions appear more evenly distributed by season than SO_2 emissions. This is primarily due to an assumed distribution of highway vehicle emissions by season, and the fact that the major sources of SO_2, which are seasonally variable, emit less NO_x.

Figure 2 shows the trend in the quantity of emissions by season on the national level. While the SO_2 percentage distribution shows a gradual change, the increase in the quantity of SO_2 emissions is more evident, especially in the summertime.

TABLE 1. PERCENTAGE DISTRIBUTION OF EMISSIONS BY SEASON

	Percentage SO_2 Emissions					Percentage NO_x Emissions			
Year	Winter	Spring	Summer	Fall	Year	Winter	Spring	Summer	Fall
1900	34.8	19.4	23.6	22.2	1900	30.8	19.9	26.1	23.2
1905	34.5	19.4	24.1	22.0	1905	31.1	19.8	26.9	23.0
1910	34.3	19.4	24.5	21.9	1910	31.7	19.8	25.8	22.6
1915	34.3	19.3	24.7	21.7	1915	31.9	19.8	25.8	22.4
1920	34.2	19.4	24.8	21.6	1920	31.6	20.2	25.8	22.4
1925	34.0	19.5	25.0	21.4	1925	30.3	20.4	26.5	22.7
1930	34.0	19.4	25.4	21.2	1930	29.5	20.7	27.0	22.9
1935	33.4	19.4	26.0	21.2	1935	28.8	20.9	27.4	22.9
1940	32.5	19.8	26.3	21.3	1940	28.8	21.2	27.3	22.7
1945	32.3	20.0	26.4	21.3	1945	28.5	21.2	27.5	22.8
1950	30.8	21.0	26.3	21.9	1950	27.6	22.3	27.0	23.0
1955	30.2	21.9	25.7	22.2	1955	27.2	23.0	26.7	23.2
1960	29.5	22.7	25.4	22.5	1960	26.5	23.4	26.5	23.5
1965	28.0	23.3	25.6	23.1	1965	25.7	23.9	26.6	23.9
1970	26.7	23.5	25.8	23.9	1970	24.6	24.1	27.2	24.2
1975	26.1	23.5	26.1	24.2	1975	24.2	24.1	27.3	24.3
1978	26.4	23.4	26.1	24.1	1978	24.5	24.0	27.2	24.3
1980	26.4	23.4	26.1	24.1	1980	24.5	24.0	27.2	24.3

Winter = December, January, February
Spring = March, April, May
Summer = June, July, August
Fall = September, October, November

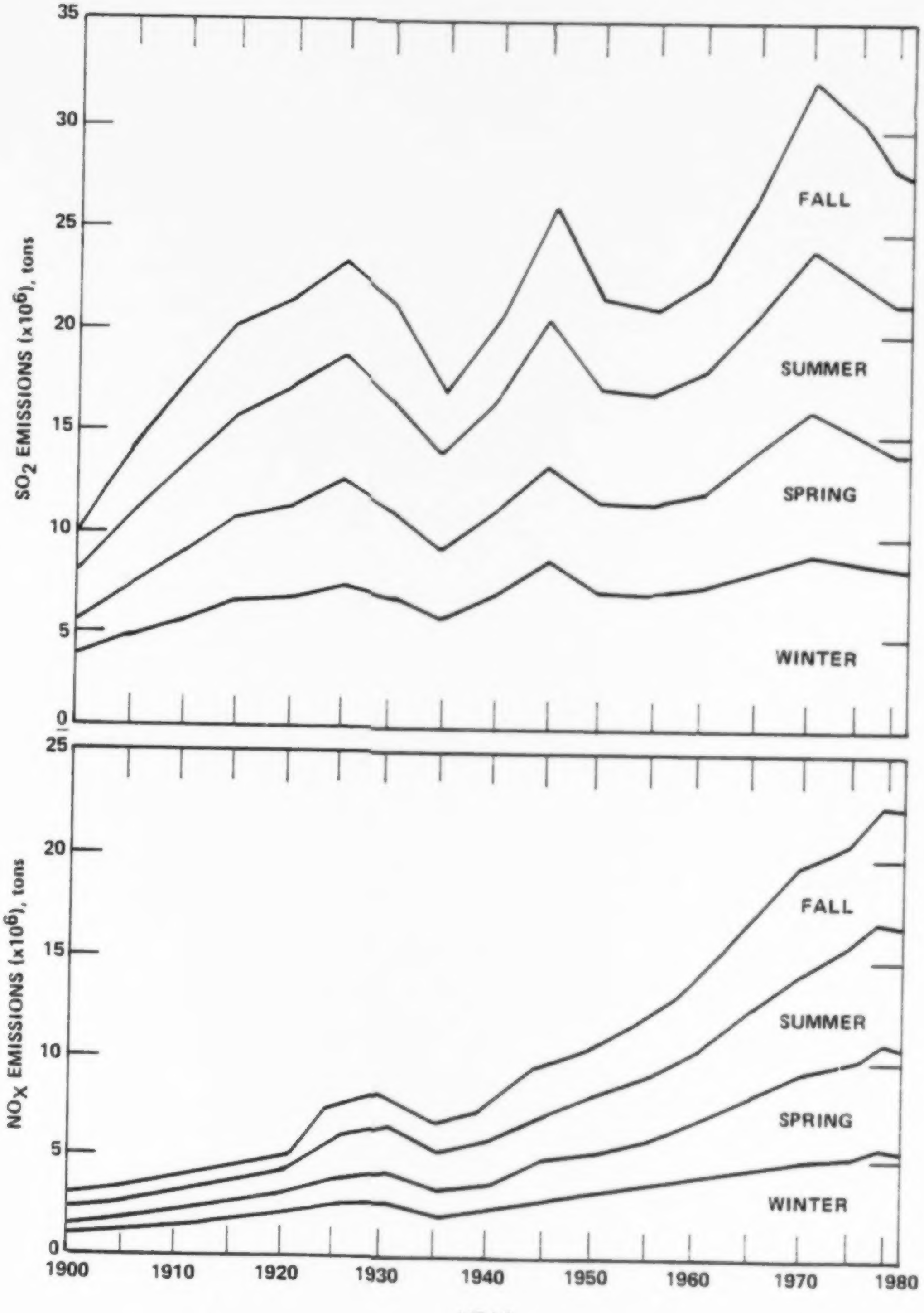

Figure 2. Total national SO$_2$ and NO$_X$ emissions by season.

CONCLUSION

The historic emission estimates presented are consistent in the estimating methodology used. They provide a basis for studying the relationship between emissions in the past and observed environmental effects including tree-ring growth patterns, material damage, and acidic deposition. The emission trends shown in this presentation should be considered the best available estimates at this time, but it should be remembered that, as work in this area continues, more refined estimates may be made. The historic emission estimates show a definite trend in terms of the total nation by stack height range and by seasonal distribution.

REFERENCES

1. G. Gschwandtner. Historic Emissions of SO_2 and NO_x Since 1900. In Proceedings: First Annual Acid Deposition Emissions Inventory Symposium, EPA-600/9-85-015 (NTIS PB 85-200004/AS), U.S. Environmental Protection Agency, Research Triangle Park, NC, pp. 38-46, May 1985.

2. G. Gschwandtner, K.C. Gschwandtner, K. Eldridge, "Historic Emissions of Sulfur and Nitrogen Oxides in the United States from 1900 to 1980," Volumes I and II. EPA-600/7-85-009a and 009b (PB 85-191 195/AS and PB 85-191 203/AS), U.S. Environmental Protection Agency, Research Triangle Park, NC, April 1985.

DEVELOPMENT OF MONTHLY EMISSIONS TRENDS FOR RECENT YEARS

by

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Development of Monthly Emissions Trends for Recent Years

D. Knudson

ABSTRACT

As atmospheric transport and deposition modeling capabilities increase and monitoring data are accumulated, it becomes desirable to also have representative emissions data. This paper describes the methodology and results of an effort to generate state-total source category-specific monthly SO_2 and NO_x emission estimates for the period 1975-1983. These data complement other data bases and support a variety of analytical efforts. The basis of the monthly emission estimates are source category-specific state-total SO_2 and NO_x emission values from the 1980 NAPAP emissions inventory. Annual emissions from the electric utility industry are allocated to monthly values using actual monthly fuel use for each year in the period. Annual emission values for other source groups were apportioned to monthly values using data which provides an indication of the monthly activity for each source group in the inventory year (1980). Annual state-total source category emissions for other years in the period were derived from the EPA emissions trends data.

The results indicate that intra-annual variability of SO_2 and NO_x emissions for each state depend on the variability of source categories contributing most to state total emissions. In states with SO_2 emissions dominated by the electric utility sector, maximum emissions occur in the summer and winter. The relative importance of these two peaks depends on such factors as climate and utility prime mover and fuel mix. Monthly NO_x emissions variations are also a function of the variability of source groups contributing the most to total state NO_x emissions. In all states, vehicle fuel consumption contributes substantially to total state NO_x emissions. Monthly variability of NO_x emissions attributable to this source category shows only small variability in most states.

DEVELOPMENT OF MONTHLY EMISSIONS TRENDS FOR RECENT YEARS

INTRODUCTION

Evaluation of long-term data on such parameters as atmospheric deposition, biological response, geochemical changes, and response characteristics of other receptor systems for trends is facilitated by appropriate emissions data for the specific period. In many cases accurate evaluation of the measured deposition and/or response of a subject receptor requires emissions data with a temporal resolution matching the critical period of the receptor. For establishing correlations and functional relationships between receptor response and emissions for trends evaluation, monthly emissions estimates are sometimes required. The data base developed in this project, therefore, complements the commitment to collection of long-term deposition and receptor system response data.

This paper describes the development and presents an overview of monthly SO_2 and NO_x emission estimates for the period 1975-1983. Review of the methodology will address the utility sector separately from all other source groups. This is because of the generally large contribution of SO_2 and NO_x emissions by the utility sector and because the methodology is more detailed for this source category. Portioning of annual state-total emissions estimates for the diverse group comprising the nonutility sector uses a number of different data bases. The selection of the appropriate monthly data base for allocation of annual emissions is a critical step in the data base development. A more thorough presentation of the methodology is provided in Reference 1.

The state-total source category-specific monthly SO_2 and NO_x estimates for all years in the period 1975-1983 are condensed into two summaries for presentation. Total regional seasonal SO_2 and NO_x values are presented for Federal Regions 1 through 6 to illustrate regional emission trends. Then, to provide an indication of differences in source category contributions to monthly emissions variability, monthly source category specific SO_2 and NO_x emissions are presented for 1980 for Illinois and New York.

METHODOLOGY

The 1980 NAPAP SO_2 and NO_x emissions inventory (version 3.0) provided state-total source category-specific emission values for SO_2 and NO_x. A six-digit level of specificity of the Source Classification Code (SCC) was used for point sources to identify the major source category (e.g., industrial boiler) activity (e.g., combustion and incineration), and fuel (coal and residual oil). For area sources, the eight digit code unique to the NAPAP inventory was used to define source groups. The apportioning of the state annual emissions used data bases that provide some measure of activity for the source category, operation, and/or fuel use on a monthly basis.

Utility Point Sources - All Years

Computation of monthly SO_2 emissions for the electric utility sector used plant emission values furnished by E.H. Pechan and Associates for the years 1975-1982[2] in combination with monthly fuel consumption reported by Energy Information Administration (EIA) Form 759 "Monthly Power Plant Report". Annual emission calculations for 1983 were performed by Argonne National Laboratory according to methodology developed by Pechan.

Computation of monthly NO_x emissions for this sector used the monthly fuel use from EIA Form 759 with plant-average NO_x emission factors. The emission factors were derived from AP-42 firing type and fuel-specific emission factors in combination with 1980 unit-level fuel use for firing of coal, lignite, gas, residual oil, and distillate oil in conventional boilers; internal combustion of natural gas and distillate oil; and gas turbines. These emission factors coincide with the fuel use categories on Form 759.

Nonutility Sources, 1980

Allocating annual SO_2 and NO_x emissions to monthly values for the diverse group of nonutility sources requires a number of equally diverse data bases. The source categories and the data bases supporting monthly allocation are defined in Table 1.

Nonutility Point and Area Sources for Other Years

Monthly SO_2 and NO_x emission estimates for nonutility source categories were derived from national emission estimates in the EPA report, "National Air Pollutant Emission Estimates: 1940-1983."[3] Values from that report for source categories matching those used for the monthly emissions estimation were normalized, with 1980 as the base year. This provided fractional national emission changes relative to 1980 for all years in the period. Therefore, the monthly source category emission fractions developed for 1980 were held constant throughout the analysis.

RESULTS

Figures 1 through 5 present seasonal total SO_2 and NO_x emissions for the period 1975 through 1983 for Federal Regions I and II, III, IV, V, and VI respectively. A characteristic apparent in all regions is the lack of seasonal variability in NO_x emissions compared to SO_2. This is probably attributable to the large contribution of vehicle fuel combustion to total NO_x emissions in most states, and the lack of variability in monthly vehicle fuel combustion.

For total SO_2 and NO_x emissions in Regions I and II (Figure 1), there is a small difference between SO_2 and NO_x emissions, with NO_x being slightly

Table 1. Data Sources for Estimation of Nonutility Monthly SO_2 and NO_x Fractions for Apportioning Annual Emissions

Description	Data Source or Approach for Estimating Monthly Fractions
Industrial external coal combustion	Quarterly Coal Report
Industrial external oil combustion	Adjusted FRB monthly production indexes
Commercial and institutional external coal combustion	Quarterly Coal Report
Commercial and institutional external oil combustion	Uniformly distributed[a]
Commercial, institutional, and industrial external combustion for space heating	Local climatological data - heating-degree-day accumulation
Commercial, institutional, and industrial internal oil combustion	Uniformly distributed[a]
Industrial chemicals: sulfuric acid, plastics, organic chemicals, explosives, carbon black, printing ink	FRB monthly production indexes for SIC 2819; inorganic chemicals
Food product processing	FRB monthly production indexes for SIC 209: miscellaneous food preparation
Primary metal: coke manufacturing, steel production, copper smelters, zinc, and other primary metals	FRB monthly production indexes for SIC 331: basic steel, coking, mill production; SIC 333-6,9: nonferrous metals
Secondary smelting: aluminum, copper lead, etc.	FRB monthly production indexes for SIC 333-6, 9: nonferrous metals
Mineral products: glass, fiberglass, gypsum products, cement, brick, pottery	FRB monthly production indexes for SIC 326: concrete and miscellaneous clay
In-process fuel use	FRB monthly production indexes for SIC 326: concrete and miscellaneous clay
Petroleum refining	FRB monthly production indexes for SIC 291: petroleum refining
Wood and paper products (including Kraft pulping)	FRB monthly production indexes for SIC 261: pulp and paper
Crude oil and natural gas extraction (including gas sweetening)	FRB monthly production indexes for SIC 131: crude oil and natural gas extraction
Solvent use	Uniformly distributed[a]
Solid waste disposal	Uniformly distributed[a]
Industrial internal gas combustion	Monthly Energy Review - natural gas used as pipeline fuel[b]

Table 1 (Cont'd)

Description	Data Source or Approach for Estimating Monthly Fractions
Incineration and open burning	Uniformly distributed[a]
Miscellaneous area sources[c]	Uniformly distributed[a]
Diesel and gasoline vehicles	1980 Highway Statistics[b]

[a] Annual data assumed to be uniformly distributed throughout the year, due to lack of monthly data.

[b] No_x only

[c] Railroad locomotives, military aircraft LTO's, civil aircraft LTO's, commercial aircraft LTO's, coal vessels, diesel oil vessels, residual oil vessels, gasoline vessels, solvent purchased, gasoline marketed, unpaved road travel, unpaved airstrip LTO's, construction, wind erosion, land tilling, forest wild fires, managed burning, agricultural burning, frost control, and structural fires.

less. SO_2 emissions have been relatively constant, while NO_x emissions are increasing slightly. Winter is the season of peak emissions for SO_2 and NO_x for the region through the entire period.

Region 3 (Figure 2) shows a small SO_2 trend downward, and no discernible NO_x trend. NO_x emissions are about 50 to 60% of regional SO_2 emissions for all years in the period. Winter is the peak season for SO_2 and NO_x for all years except 1983, in which summer is the peak emission season.

Region IV shows a steady decline in SO_2 emissions from a maximum in 1977. There is little change in NO_x emissions through the period, with NO_x being about 50% of the SO_2 emissions.

In Region V, SO_2 emissions have decreased steadily since 1977 to a value about double the NO_x emissions in 1983. NO_x emissions have changed little throughout the period. Winter is the peak season for NO_x and SO_2 emissions for all years in the period.

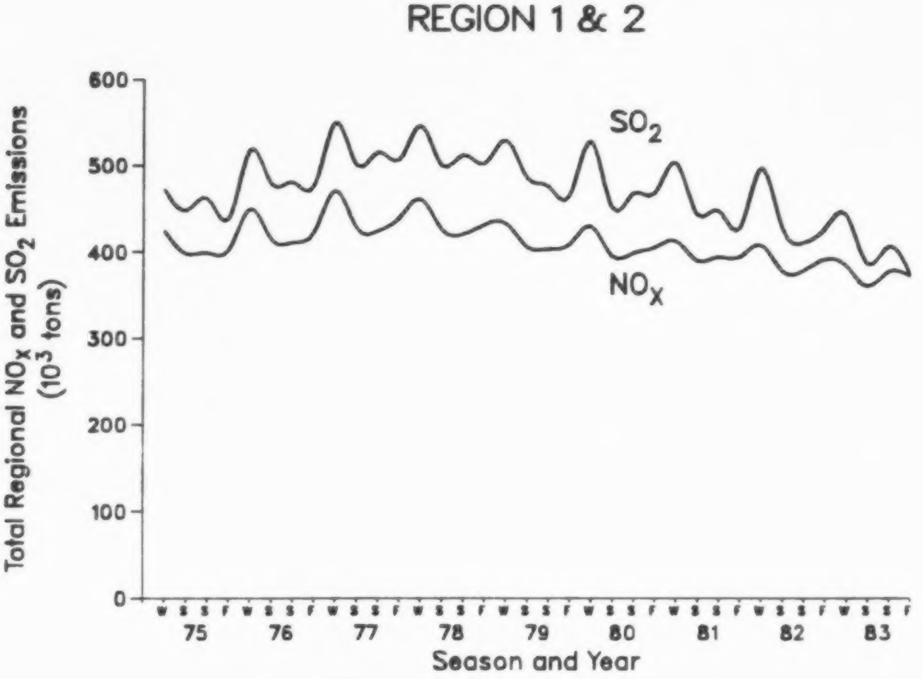


Figure 1 Regions 1 and 2 Seasonal SO_2 and NO_x Emission Totals for the Period 1975-1983.

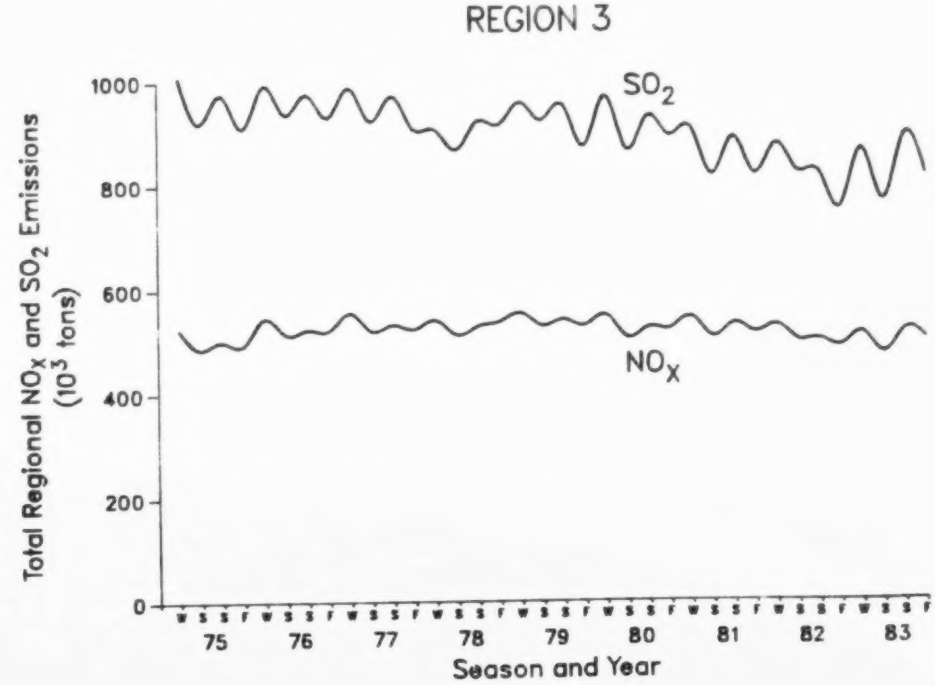


Figure 2 Region 3 Seasonal SO_2 and NO_x Emission Totals for the Period 1975-1983.

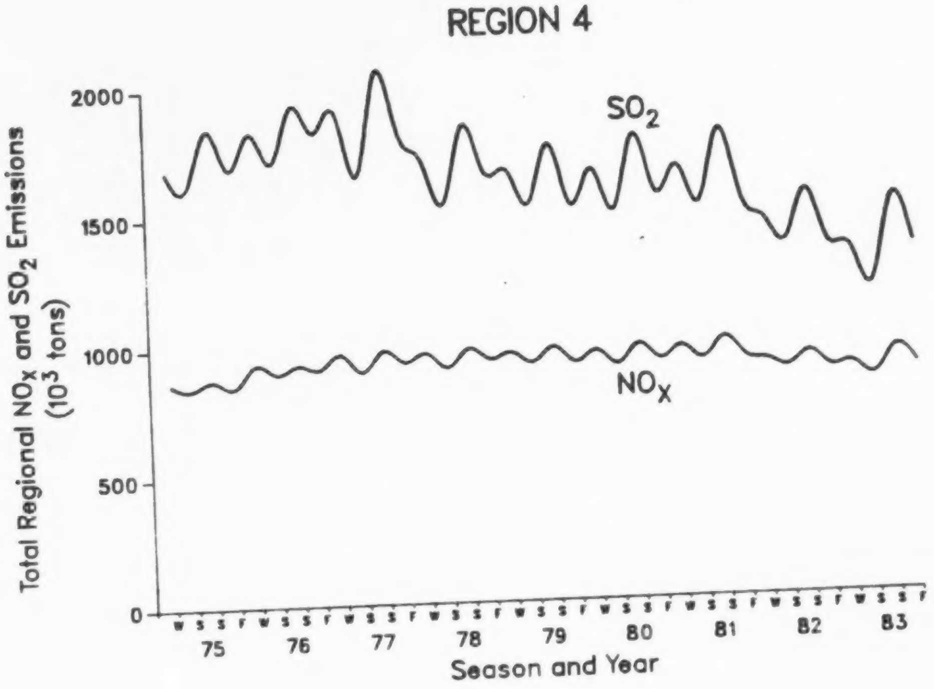


Figure 3 Region 4 Seasonal SO$_2$ and NO$_x$ Emission Totals for the Period 1975-1983.

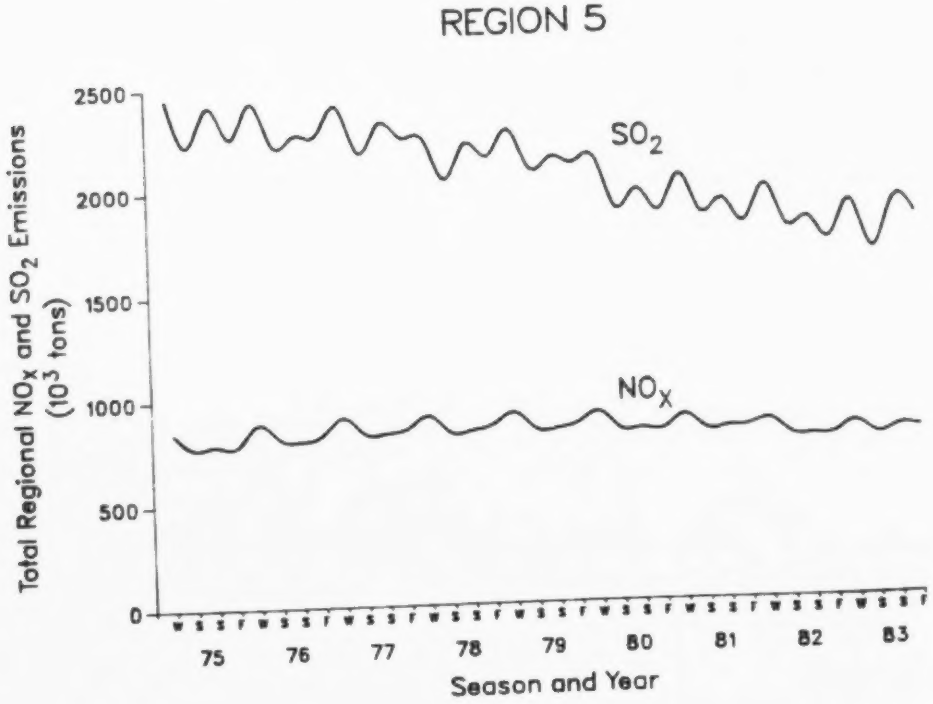


Figure 4 Region 5 Seasonal SO$_2$ and NO$_x$ Emission Totals for the Period 1975-1983.

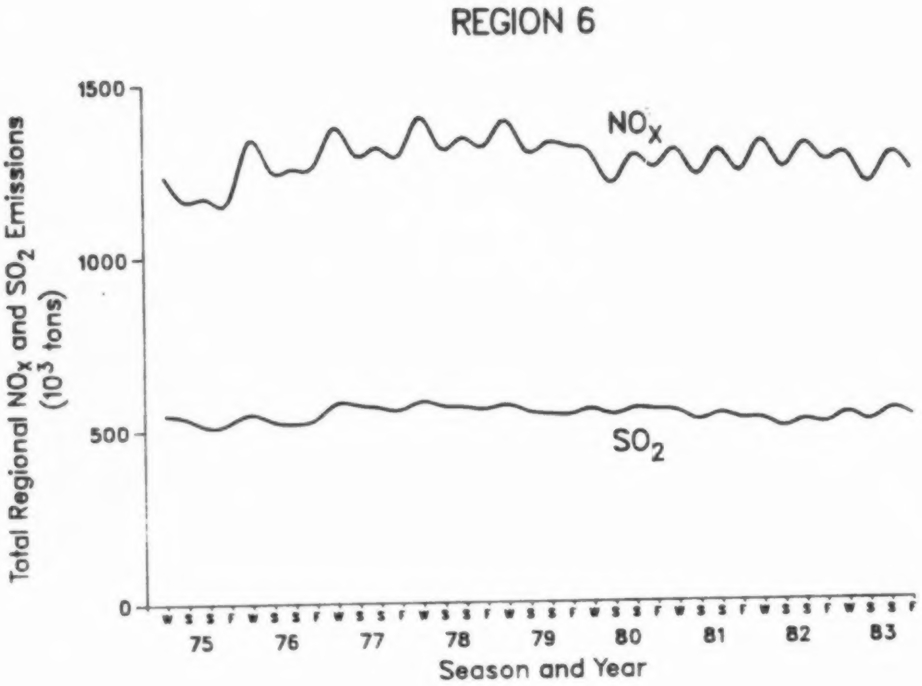
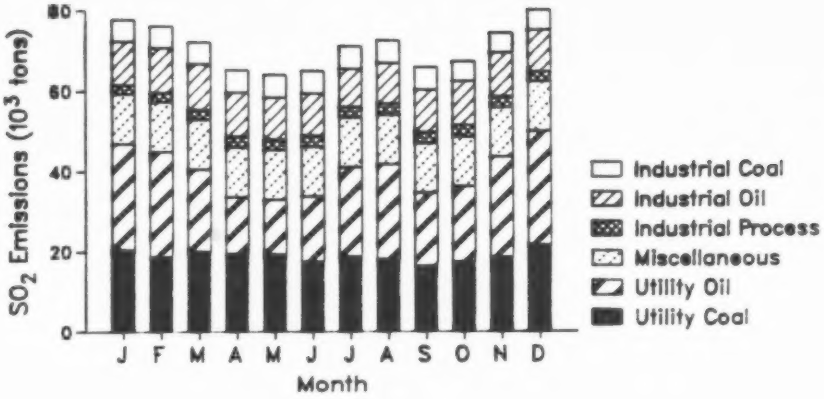


Figure 5 Region 6 Seasonal SO_2 and NO_x Emission Totals for the Period 1975-1983.

New York



Illinois

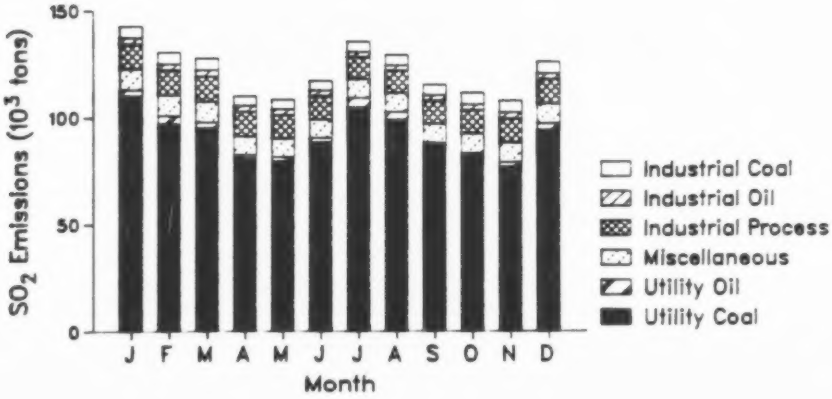
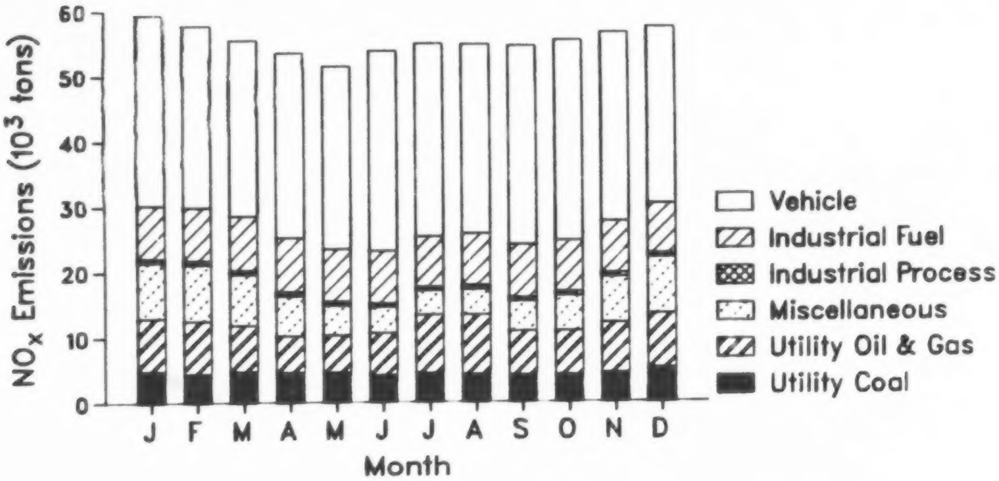


Figure 6 Monthly 1980 SO_2 Emissions by Source Category for New York and Illinois.

New York



Illinois

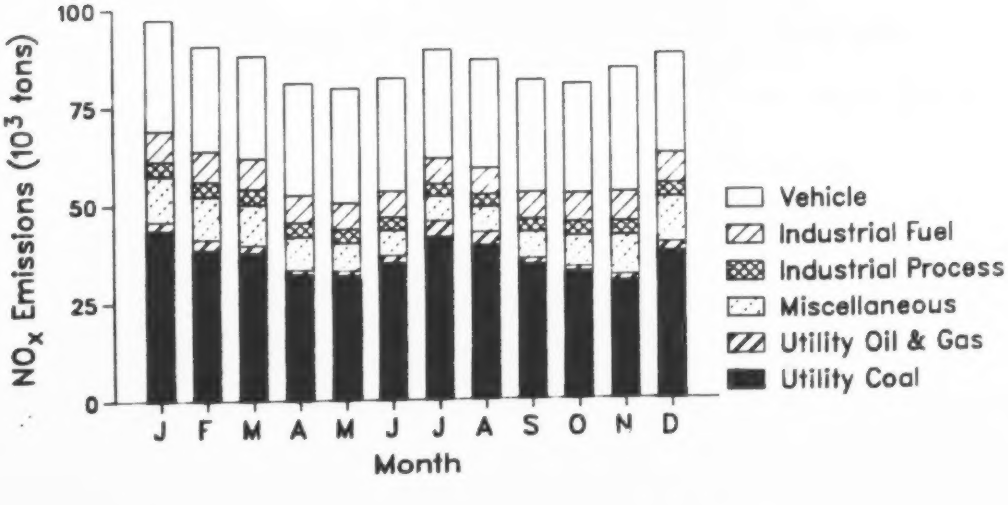


Figure 7 Monthly 1980 NO$_x$ Emissions by Source Category for New York and Illinois.

In Region VI, NO_x emissions are roughly a factor of two higher than SO_2 emissions. Neither SO_2 nor NO_x emissions have changed significantly over the period of interest. There is a slight winter peak for both pollutants for all years except 1983, for which summer is the season for maximum SO_2 emissions.

Figures 6 and 7 present 1980 monthly source category SO_2 and NO_x emission estimates for Illinois and New York, respectively. In both states the utility sector contributes the most to SO_2 emissions. In Illinois monthly variability in utility coal combustion dominates the total state monthly SO_2 emissions variations. In comparison, utility coal derived SO_2 emissions for New York have little variability through the year. The variations in total New York state SO_2 emissions is largely attributable to utility oil combustion fluctuations. In New York, source categories defined as industrial oil and miscellaneous also have substantial contributions to total state SO_2 emissions, but show little monthly variability in SO_2 emission rates.

Monthly emissions of NO_x also show differences between the two states (Figure 7). NO_x emissions in Illinois are clearly dominated by utility coal combustion and vehicle fuel use. The vehicle source category is the overwhelming contributor to NO_x emissions in New York State. However, coincident minimum monthly NO_x emissions for the utility oil, miscellaneous, and industrial oil combustion categories combine to yield a state-total minimum NO_x emission rate in the late spring to early summer period, even though vehicle-related emissions are not at their minimum during these months.

CONCLUSIONS

Monthly state-total SO_2 and NO_x emissions by source category for the years 1975-1983 have been prepared. The allocation of annual emissions to monthly values used a diverse set of data describing source category-specific fuel use or related activity. Definition of the monthly data base used for allocation is the critical step in the development of the monthly data.

General regional patterns of monthly SO_2 and NO_x emissions are dependent on the characteristics of each region. In the northeast (Regions I-III), SO_2 emissions have decreased slightly while NO_x emissions have been increasing through the 1975-1983 period. In these regions winter is the peak SO_2 and NO_x emission season. Region IV has experienced a significant decline in SO_2 emissions and little to no increase in NO_x, with summer being the season of maximum emissions for both pollutants. Region V has shown the largest decrease in SO_2 emissions, with little change in NO_x emissions across the period. Winter is the season of maximum SO_2 and NO_x emissions for Region V. The relative portions and trends in SO_2 and NO_x emissions for Region VI are distinctively different from the patterns in the eastern regions. In Region VI, NO_x emissions are roughly a factor of two higher than SO_2 emissions, with neither showing any trend through the period. Unexpectedly for Region VI, winter is the season of peak SO_2 and NO_x emissions for all years in the period, except 1983. In 1983, SO_2 emissions peaked in the summer.

Generalizations of state-level monthly emissions variability are impossible to make. The pattern of monthly SO_2 and NO_x emissions depends on the relative contributions of source categories in each state and their monthly emission pattern.

ACKNOWLEDGEMENT

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REFERENCES

1. Knudson, D.A. "An Inventory of Monthly Sulfur Dioxide Emissions for the Years 1975-1983," ANL/EES-TM-277 (April 1985).

2. E.H. Pechan and Associates, "Estimates of Sulfur Dioxide Emissions from the Electric Utility Industry," EPA 600/7-82-061a and b (November 1982).

3. U.S. EPA "National Air Pollutant Emission Estimates, 1940-1983," EPA 450/4-84-028 (December 1984).

APPLICATIONS OF EMISSIONS DATA IN THE NAPAP ASSESSMENT REPORT

Paul Schwengels
Office of Air and Radiation
U.S. Environmental Protection Agency

Presented at:

Second Annual Acid Deposition Emission Inventory Symposium

Charleston, South Carolina

November 12-14, 1985

APPLICATIONS OF EMISSIONS DATA IN THE NAPAP ASSESSMENT REPORT

By: Paul Schwengels
Office of Air and Radiation
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ABSTRACT

A great deal of emissions data has been assembled for use in the forthcoming NAPAP Assessment Report. This includes natural and man-made sources of a number of relevant chemical species or classes in both the United States and Canada. Emissions data will be used in the report in several ways. First, an emissions section will summarize best available information on substances of interest, compare natural and man-made levels, provide regional distributions and comparisons, and evaluate national and regional trends in man-made emissions where possible. In addition, emissions data have been used to support analyses and modeling for the atmospheric processed and materials damage sections of the report. This paper briefly describes the various data sets which have been assembled to meet these different needs in the assessment. It identifies the different types of data needed to support different types of analyses or comparisons. Uncertainties and limitations in available data and their impact on assessment will be reviewed, as well as data improvements needed to support improved assessments in the future. Some preliminary approaches to analysis and presentation of data being considered for the Assessment Report will also be discussed.

Note: The text of this paper was not available at the time of publication of the proceedings.

OVERVIEW OF THE DEVELOPMENT OF THE 1985 NAPAP EMISSIONS INVENTORY

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Presented at:

Second Annual Acid Deposition Emission Inventory Symposium

Charleston, SC

November 12-14, 1985

OVERVIEW OF THE DEVELOPMENT OF THE 1985 NAPAP EMISSIONS INVENTORY

ABSTRACT

The National Acid Precipitation Assessment Program (NAPAP) needs a current and detailed emissions inventory to support its activities. To meet this objective, NAPAP and the United States Environmental Protection Agency (EPA) have selected 1985 as the base year for the inventory. Data collected by the States during 1985 for the National Emissions Data System (NEDS) will be used to develop the NAPAP emissions inventory. The NEDS was selected to permit the design of a data collection program that will meet NAPAP's requirements for both quality assurance and spatial, temporal, and species resolution. Ensuring the accuracy of more than 4 million items of data that the States will collect for NEDS is a challenging task. To meet this challenge, EPA has developed a five part management strategy. This strategy emphasizes the high priority of the NEDS in 1985, establishes EPA and State accountability for the NEDS data, delineates quality assurance procedures for the data, provides financial assistance to States that require it, and ensures that participating State and EPA personnel clearly understand the NEDS data requirements. This management strategy will result in the development of NEDS data that will have unparalleled quality.

INTRODUCTION

The National Acid Precipitation Assessment Program (NAPAP) requires emissions data for three reasons: to support atmospheric modeling, to explore relationships between sources of emissions and receptors of acid deposition, and to facilitate examination of regulatory alternatives. The reliability of NAPAP's activities in these areas can be no better than the data on which they are based. Thus, to produce reliable results, these activities must be founded on the development of a comprehensive emissions inventory with a high degree of quality control and of species, temporal, and spatial resolution.

As illustrated in Figure 1, the basis for NAPAP's estimates of anthropogenic emissions will be the 1985 emissions inventory conducted by the States for the National Emissions Data System (NEDS). This system is maintained by the National Air Data Branch of the U.S. Environmental Protection Agency (EPA). The goal of this project is to ensure the best ever development of a NEDS data base to meet NAPAP's needs for 1985 emission inventory data.

DISCUSSION

The NEDS is a computerized system which accepts, stores, and reports source data and emissions data for both point and area sources. Point sources are considered to be stationary sources of pollution that emit at least 100 tons annually of particulate matter, sulfur dioxide, nitrogen oxides, or reactive volatile organic hydrocarbons, or that emit at least 1000 tons annually of carbon monoxide. Area sources are considered to be stationary sources with emission rates below these thresholds and all mobile sources of particulate matter, sulfur dioxide, nitrogen oxides, reactive volatile organic hydrocarbons, or carbon monoxide.

Point source data are collected by both State and local air pollution control agencies. The data are obtained for each source at an individual plant through questionnaires, visits, and emission tests. For an average plant, a State must collect, check, and report 350 items of data. After conducting quality assurance checks on the data, States enter the data onto forms or computer tapes compatible with NEDS and transmit the data to their EPA Regional Offices. States are required to make an annual report to EPA for all of the sources meeting the above criteria.

Area source data are developed by EPA's National Air Data Branch. Development of the area source data relies upon published information for calculating area source activity levels, such as quantities of fuels distributed to States, vehicle-miles travelled by motor vehicles and so forth. State level data are allocated to counties using employment, population, or similar statistics. Emissions are calculated using emission factors from standard EPA references such as AP-42 and the MOBILE3 highway vehicle emission factor model.

The major thrust of the 1985 data collection effort is to ensure a high quality, timely point source data submittal by the States. In deciding to use 1985 NEDS emissions data for NAPAP's purposes, EPA recognized that careful management would be needed to superimpose NAPAP's data requirements on the State emissions inventory process. The management strategy that EPA has developed to help the States meet NAPAP's requirements consists of five elements:

- Emphasizing priority: emphasizing the importance of meeting the NAPAP data requirements to managers in State offices and EPA Regional Offices;
- Establishing accountability: establishing a network of managers in the State offices and EPA Regional Offices that will be responsible for the NEDS data;
- Ensuring accuracy: establishing quality control procedures for preparation and submittal of the source and emissions data;

- Assisting States: pinpointing and eliminating any State resource constraints; and

- Establishing communication: ensuring that all participating State and EPA personnel understand the NAPAP requirements for the emissions data to be collected.

The first element of the strategy was to establish the priority for the 1985 State NEDS inventory. This has been accomplished through a memorandum from EPA Administrator Lee Thomas to State Air Program Directors and to EPA Regional Administrators. This letter urged State and EPA Regional assistance to produce an accurate and comprehensive emissions inventory. The letter also emphasized the high priority that EPA attached to these goals. To focus additional attention on these goals within EPA, the Administrator's Strategic Planning and Management System for fiscal year 1986 includes NEDS performance milestones for managers in the Regional Offices.

The second element of the strategy was to establish accountability. A network of managers responsible for ensuring the quality and scheduling of the emissions data was established during August and September 1985. This network will consist of individuals in both Regional Offices and State Air Program Offices. Accountability for EPA managers is further emphasized through the performance milestones mentioned above.

The third element of the strategy is the most challenging one. Ensuring the accuracy of more than 4 million items of source and emissions data that will be collected during 1985 is a difficult task under the best of circumstances. Given the limited resources typically encountered in the State air program offices, the task of selecting appropriate quality assurance procedures is made even more complex. The quality assurance procedures that NAPAP and EPA are to employ attempt to strike a balance between vigorous investigations and limited State resources. States and EPA Regional Offices will be provided with written guidance concerning the quality assurance checks that NAPAP will conduct. This will not only focus attention both on critical elements in the NEDS data and on appropriate calculation procedures, but will also minimize errors. Also, Regional Offices will be provided with copies of computer programs designed to check the data submitted by the States for potential errors. When questions about the data are raised, the Regional Offices will contact the States to resolve any problems. Finally, NAPAP will perform its quality assurance checks on high-priority data items to ensure that errors have been corrected by the State and Regional Office efforts. NAPAP will also cross-check the data received with independent data sources, such as U.S. Department of Energy data for individual electric utility plants.

The fourth element of the strategy is assisting the States. EPA has allocated $500,000 from Clean Air Act Section 105 air quality grants to help States that need extra resources. A similar amount of assistance as contract support is to be provided from NAPAP funds. The resource assistance plan is discussed in detail in the paper by David Johnson, titled Assistance to States in the Development of the 1985 NAPAP Emission Inventory.

The final element of the management strategy is communication. On July 30, 1985, OAQPS transmitted written guidance to each EPA Regional Office Air Program Director which discussed three initiatives being taken to ensure that the network of State and EPA personnel responsible for the NEDS emissions data understand both their roles and the data collection requirements. These initiatives include the organization of telephone conferences to coordinate work between EPA personnel in OAQPS and in the Regional Offices, and the preparation of written quality assurance guidance for EPA and State personnel. A workshop for State and Regional project managers was held on October 24 and 25. OAQPS will maintain contact with the Regional and State project managers throughout the remainder of the project to follow up on specific problems that develop.

Work is also planned to improve the methodology for calculation of NEDS area source emissions to ensure that all significant source categories of emissions are accounted for. Development of an improved methodology for highway vehicles is also anticipated. This is necessary to improve the accuracy of the emissions estimates for this source category which accounts for about one-third of national NO_x and VOC emissions. The new methodology will produce better estimates of vehicle-miles travelled to be used with MOBILE3 emission factors to calculate emissions.

Finally, the NEDS point and area source data developed by the States and NADB will be processed by NAPAP to produce the spatial, temporal, and species resolution of emissions needed by NAPAP. The procedures for accomplishing this have been described by Fred Sellars in the paper titled Development of an Emissions Inventory to Support Testing of the Eulerian Regional Acid Deposition Model.

CONCLUSION

The management strategy developed by EPA represents a strong commitment to achieving NAPAP's goals for a 1985 emissions inventory. This effort has the highest priority of any emission inventory project conducted since the initial development of NEDS in the early 1970's. To achieve the desired goals will require an intensive and coordinated effort by the States and EPA. The expectation is that the inventory data developed will be the best ever during the time NEDS has been in existence.

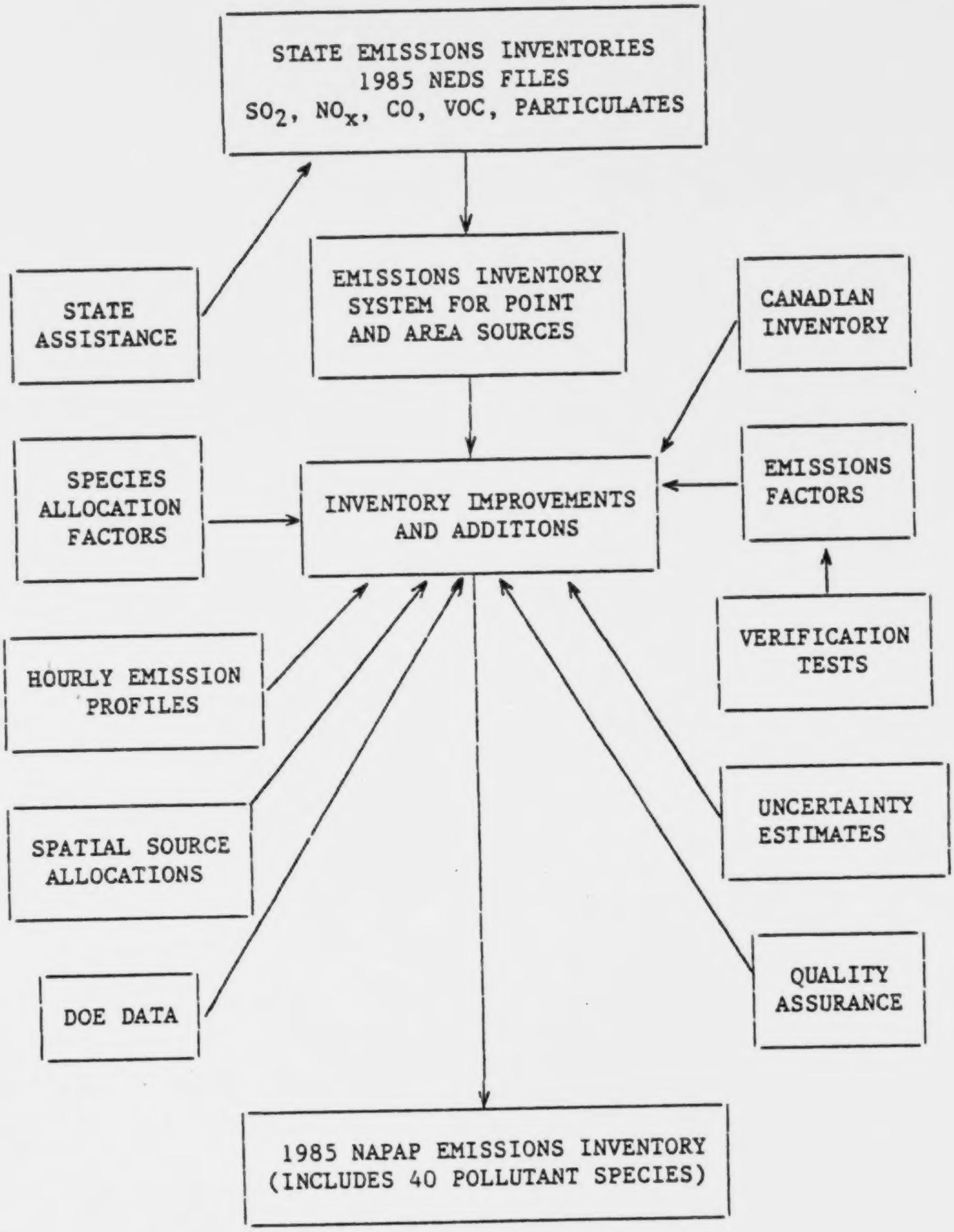
STATE EMISSIONS INVENTORIES
1985 NEDS FILES
SO_2, NO_x, CO, VOC, PARTICULATES
STATE ASSISTANCE
EMISSIONS INVENTORY SYSTEM FOR POINT AND AREA SOURCES
CANADIAN INVENTORY
SPECIES ALLOCATION FACTORS
INVENTORY IMPROVEMENTS AND ADDITIONS
EMISSIONS FACTORS
VERIFICATION TESTS
HOURLY EMISSION PROFILES
UNCERTAINTY ESTIMATES
SPATIAL SOURCE ALLOCATIONS
DOE DATA
QUALITY ASSURANCE
1985 NAPAP EMISSIONS INVENTORY
(INCLUDES 40 POLLUTANT SPECIES)

Figure 1. Relationship Between State and NAPAP Emissions Inventories for the 1985 Base Year

ASSISTANCE TO STATES IN THE
DEVELOPMENT OF THE 1985
NAPAP EMISSION INVENTORY

by

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EPA Contract No.

EPA Project Officer:

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Presented at:

Second Annual Acid Deposition Emission Inventory Symposium
Charleston, South Carolina

November 12-14, 1985

ABSTRACT

The Environmental Protection Agency (EPA) is working with the State and Territorial Air Pollution Program Administrators (STAPPA) Acid Rain Committee to identify the capabilities and needs of States with regard to development of the 1985 National Acid Precipitation Assessment Program (NAPAP) emission inventory. A survey has been distributed to the States asking them to determine what level of inventory effort they could provide for 1985 given their current resources and activities and what types and levels of support are needed to ensure the desired information can be collected and submitted. The results of the survey will be used in determining what type of assistance will be provided with the additional funds available for this project.

Introduction

The 1985 NAPAP emission inventory effort will call on States to make a concerted effort to collect and submit emission-related data. However, since the emission inventory development and maintenance activities and capabilities vary across the States, it is necessary to determine what assistance or support might be needed to enable all States to produce consistent and high-quality emission inventory information. Once these needs are determined, an appropriate strategy to provide the assistance can be developed based on program goals and available resources.

Background

In June 1985, the Administrator of the U.S. Environmental Protection Agency (EPA) wrote to each State Environmental Director explaining the importance of the 1985 emission inventory and the role of States in the development of the inventory. The Administrator said that he was giving the 1985 inventory project a high priority within EPA, and he urged the State Directors to also give the project high priority in their States. The Administrator also commented that he realized that the inventory development activities and capabilities varied across the States and that the critical needs associated with producing consistent and high-quality emission inventories should be identified. Once these needs were identified, ways of satisfying these needs and solving major problems should be determined.

The Administrator stressed his desire to work closely with the States in determining their inventory capabilities and needs. He stated that EPA's interface with the States would be through the STAPPA Acid Rain Committee, chaired by Mr. Jim Hambright (head of the Pennsylvania air program). Staff of EPA first met with the STAPPA committee in April 1985 to discuss the objectives of the 1985 inventory project and how EPA and STAPPA could work together to identify and address State inventory needs and concerns. The STAPPA committee and EPA agreed that a greater effort within the existing National Emission Data System (NEDS) context could produce a worthwhile improvement in emission inventory information and that an appropriate way to determine the capabilities and needs of States would be through a survey.

A survey was developed and mailed by STAPPA to the 48 contiguous States and the District of Columbia in July 1985. The primary goals of the survey were to determine (1) what level of inventory effort the States could provide for 1985, given their current inventory activities and resources and (2) what types and levels of support were needed to ensure the States could provide the desired emission inventory information.

Results of the STAPPA Emission Inventory Survey

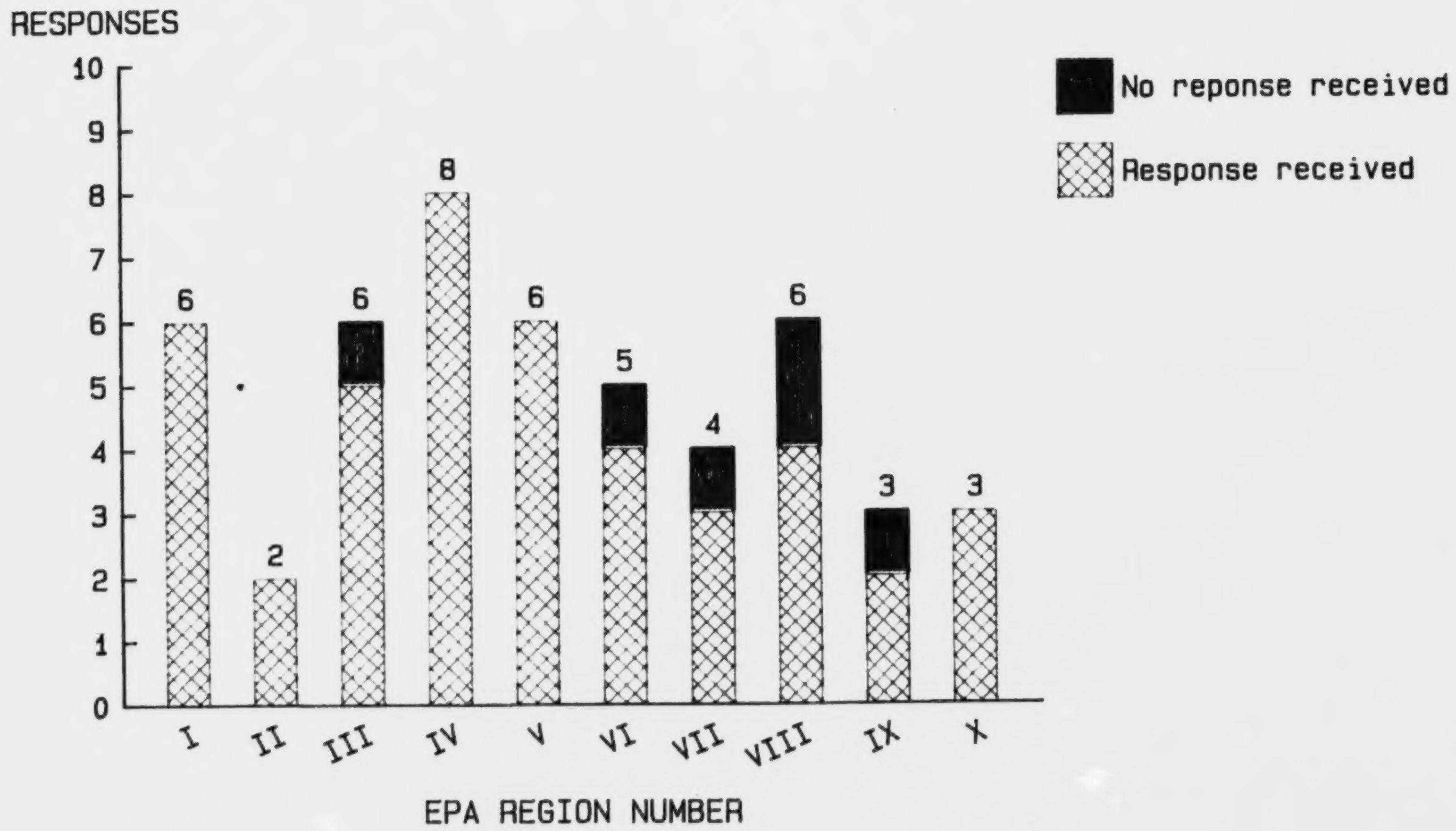
The survey drew an effective and widespread response from the States. Out of the 49 surveys that were mailed, 42 had been returned by October 17, 1985. Figure 1 shows the response in each EPA Region, which illustrates an effective response to the survey across the country. It was also evident that many States gave considerable time and effort in preparing their responses, with some States providing detailed estimates of man-hour and material needs associated with the 1985 effort. The following discussion summarizes the results of the survey.

Inventory Capabilities With Current Resources - The first primary question of the survey asked whether States would be able to provide by July 1, 1986, the 1985 emission data for various sources given the States' current inventory activities and resources. States were asked to specify whether they would be able to provide emission data needed for (a) all, (b) more than half, (c) less than half, or (d) none of the sources grouped by pollutant and size categories. The responses of the States are summarized in Figures 2-6. Figure 2 illustrates that over two-thirds of the States could provide the emission-related information for all sulfur dioxide (SO_2) sources greater than 2,500 tons per year (TPY) with current resources; over half could provide the data for SO_2 sources greater than 1,000 TPY; and over a third could provide the data for all SO_2 sources greater than 100 TPY without needing additional assistance. The most significant change in the distribution occurs when the size cutoff is lowered to 100 TPY. Considerably fewer States are able to provide data for all sources emitting more than 100 TPY of SO_2, and most States could provide the data for only a portion or none of their sources.

Figure 3 illustrates an almost identical pattern in the capability of States regarding sources of oxides of nitrogen (NO_x). The similarity in Figures 2 and 3 shows an expected pattern, which is apparent also for sources of volatile organic compounds (VOC's), particulate matter (PM), and carbon monoxide (CO), that reflects the increasing demands on State resources as the size cut-off becomes smaller. However, the similarity in a State's capabilities regarding SO_2 and NO_x sources also results from the considerable overlap in sources; that is, most sources of SO_2 emissions are also sources of NO_x and vice versa.

Figure 4 shows the capabilities of States to provide the emission data for VOC sources. Figures 5 and 6 show the capabilities of the States to provide the emissions data for sources of particulate and carbon monoxide. Although the patterns in the capability of States to provide data for these sources are quite similar to the patterns illustrated in Figures 2 and 3 for SO_2 and NO_x, across the board there are fewer States capable of providing data for all sources of VOC, PM, and CO in the particular size categories.

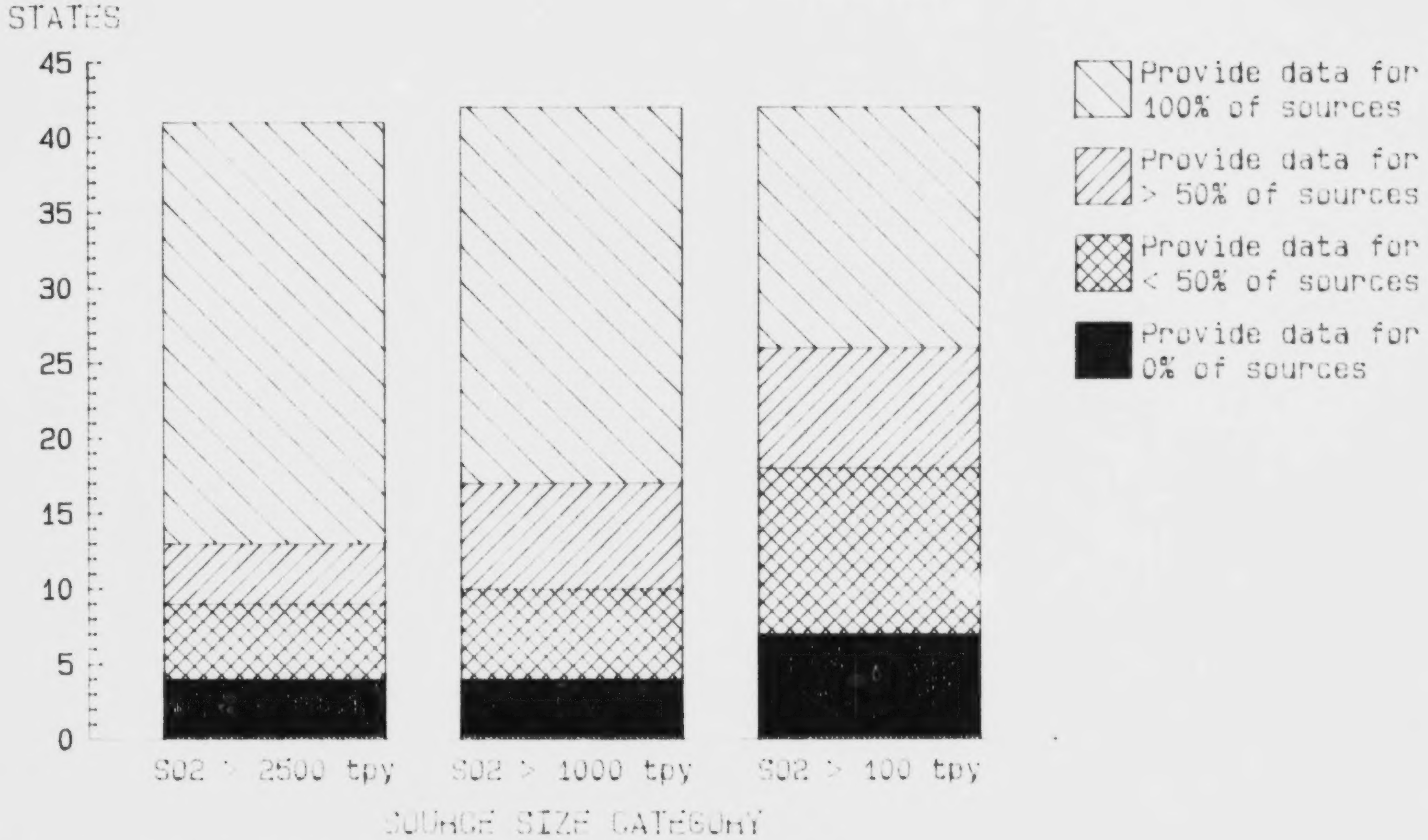
Figure 1. SURVEY RESPONSES RECEIVED
1985 NAPAP EMISSIONS INVENTORY



Based upon 42 of 49 possible responses.

October 30, 1985

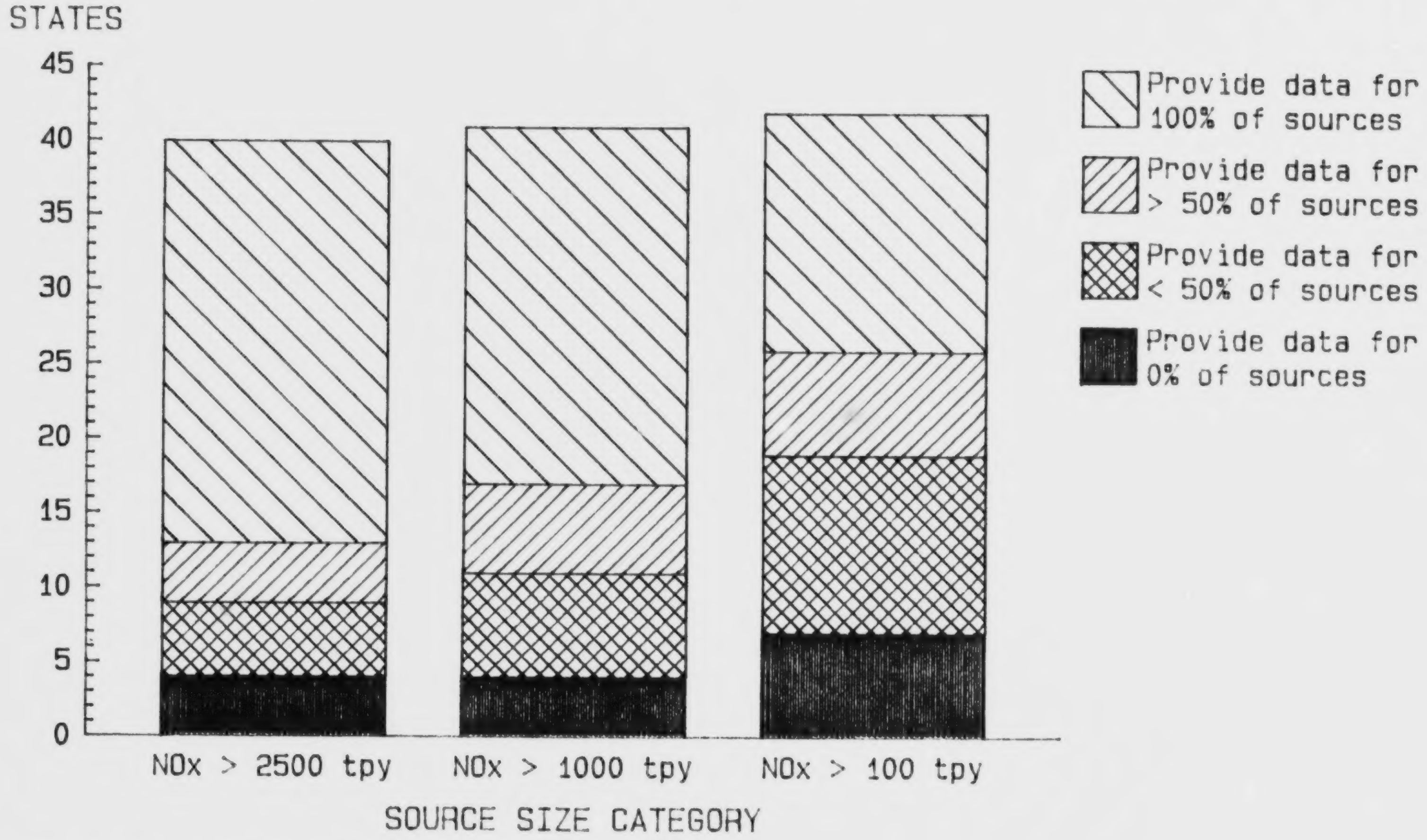
Figure 2. ABILITY TO PROVIDE 1985 EMISSIONS DATA FOR SO2 SOURCES (NAPAP Emissions Inventory)



Based upon 42 of 49 possible responses.

October 30, 1985

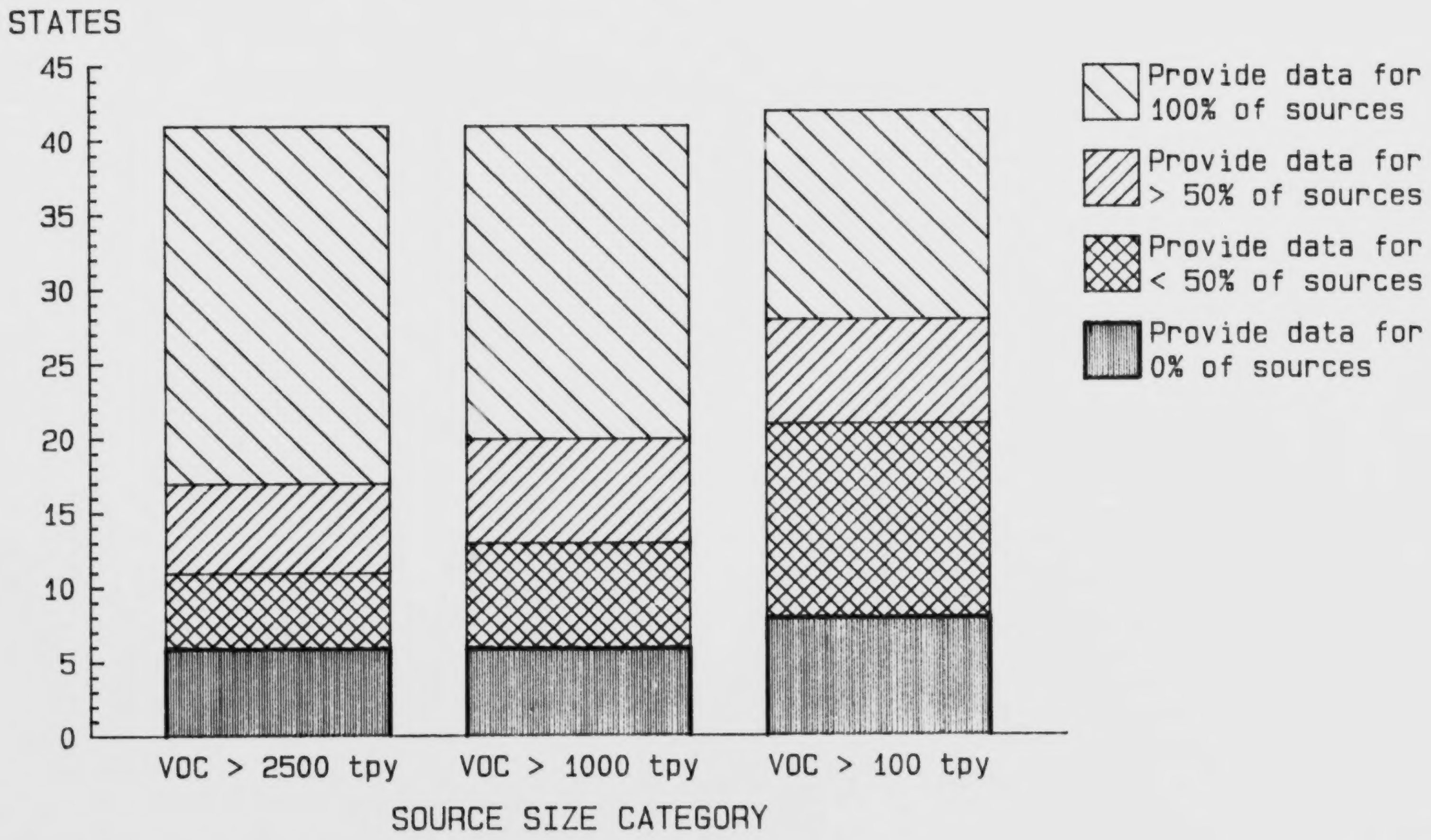
Figure 3. ABILITY TO PROVIDE 1985 EMISSIONS DATA FOR NOx SOURCES
(NAPAP Emissions Inventory)



Based upon 42 of 49 total possible responses

October 30, 1985

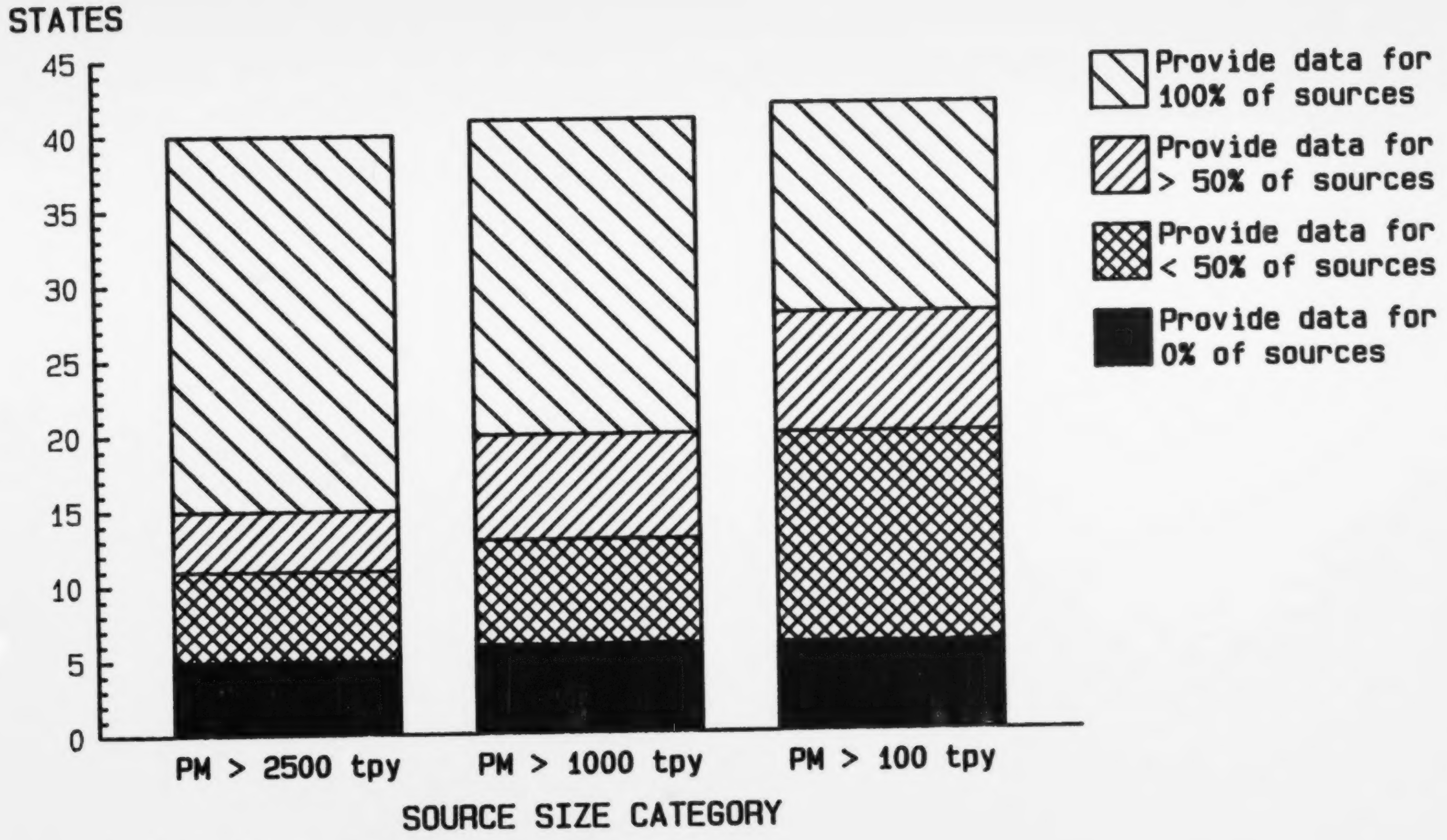
Figure 4. ABILITY TO PROVIDE 1985 EMISSIONS DATA FOR VOC SOURCES
(NAPAP Emissions Inventory)



Based upon 42 of 49 total possible responses.

October 30, 1985

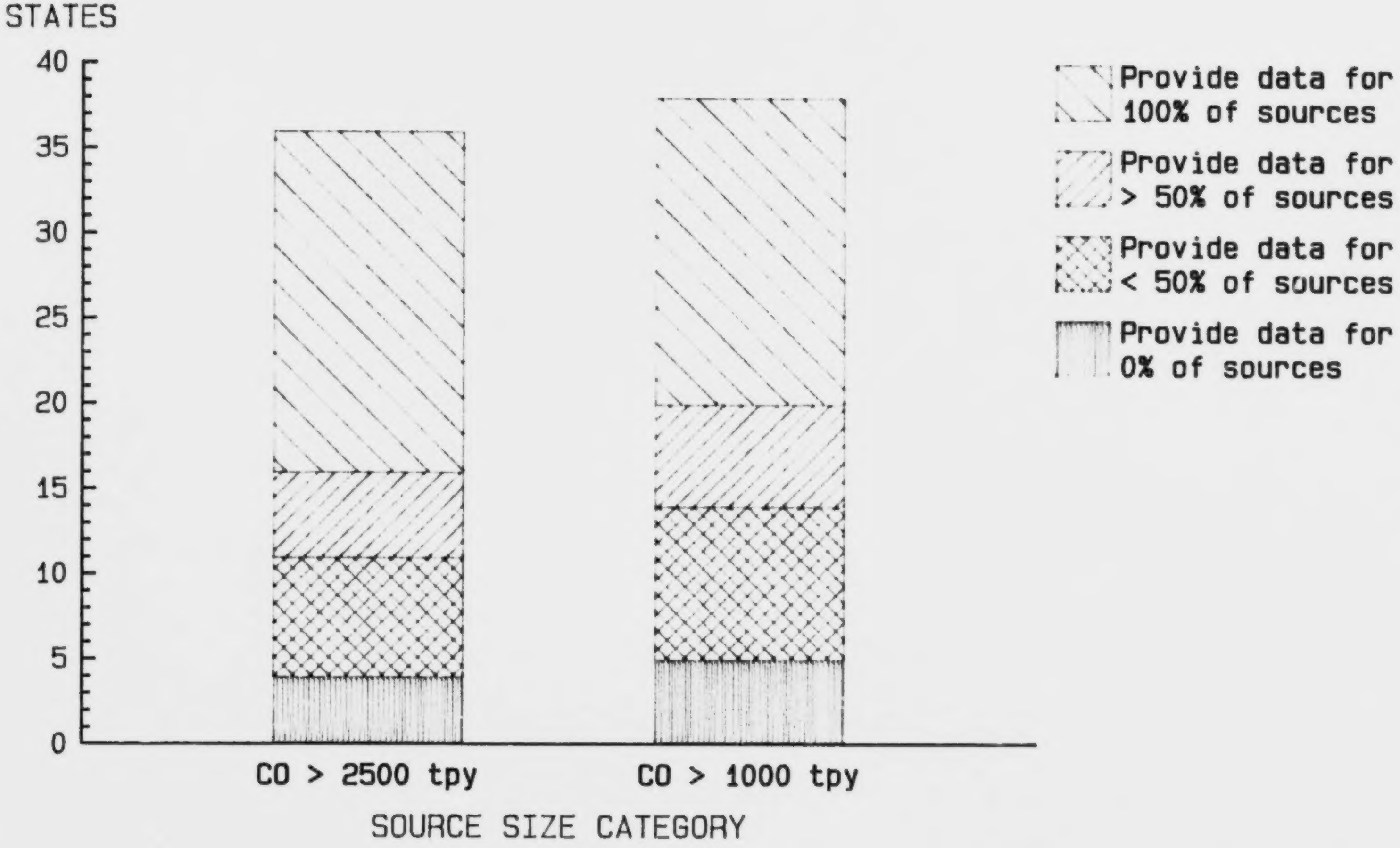
Figure 5. ABILITY TO PROVIDE 1985 EMISSIONS DATA FOR PARTICULATE MATTER SOURCES (NAPAP Emissions Inventory)



Based upon 42 of 49 total possible responses.

October 30, 1985

Figure 6. ABILITY TO PROVIDE 1985 EMISSIONS DATA FOR CARBON MONOXIDE (CO) SOURCES (NAPAP Emissions Inventory)



Based upon responses from 40 of 49 total possible responses.

It should be noted that these figures are based on the number of States responding a particular way to a specific question. If the number of plants, emission points, or total emissions from various source categories were considered, the implications for resource allocation might be different. For example, Figure 7 illustrates the number of sources and emission points associated with the States' ability to provide emission data for sources emitting more than 2,500 TPY of SO_2. In this case, considering either the number of State responses or the associated plants or points would seem to have the same implications for the coverage of emission sources. However, for NO_x (see Figure 8), the implications are different depending on whether State responses or number of plants or points are used. Still, neither of these figures show what portion of the total inventory would be accounted for, which could have other implications for resource allocation. All of these variables will need to be considered as further evaluation of the surveys occurs.

In addition to being asked about their being able to provide the desired emission information, States were asked whether they could also confirm the emission estimates with their SO_2 and NO_x sources. Figure 9 illustrates how many States could confirm the emission estimates they would be able to develop for sources emitting more than 2,500 TPY of SO_2. For example, of the 26 States that would be able to develop emission estimates for all sources emitting over 2,500 TPY of SO_2, 21 would be able to confirm those estimates and five States would be able to confirm the estimates of only a portion or none of those sources. Figure 10 illustrates similar confirmation capabilities regarding SO_2 sources emitting more than 1,000 TPY. The responses regarding NO_x sources were similar. One indication of these figures is that almost all States that would be able to estimate emissions for all their sources would be able to confirm those emissions, whereas those States who could estimate emissions for only a portion of their sources would be able to confirm emissions for only a portion of those sources.

Amount and Type of Assistance Needed - If a State would need assistance in providing the 1985 emission inventory information that was needed, it was asked to determine the amount of assistance (in man-hours and material costs) that would be needed to (a) develop and confirm emission estimates for sources emitting more than 2,500 TPY of SO_2 or NO_x: (b) develop emission estimates for sources emitting more than 100 TPY of SO_2, NO_x, or VOC; and (c) develop emission estimates for sources emitting more than 100 TPY of PM or 1,000 TPY of CO. Figure 11 presents the total requested assistance associated with these three levels of inventory development. Of the total $2.05 million requested, about 20 percent or $410,000 would be needed to develop and confirm emission estimates for sources emitting more than 2,500 TPY of SO_2 or NO_x. Over half of the amount requested would be used to develop the emission estimates for sources emitting more than 100 TPY of SO_2, NO_x, or VOC, and about a fourth would be needed for the sources of PM and CO.

Figure 7. ABILITY TO PROVIDE 1985 EMISSIONS FOR SO2 SOURCES >2500 TPY (NAPAP Emissions Inventory)

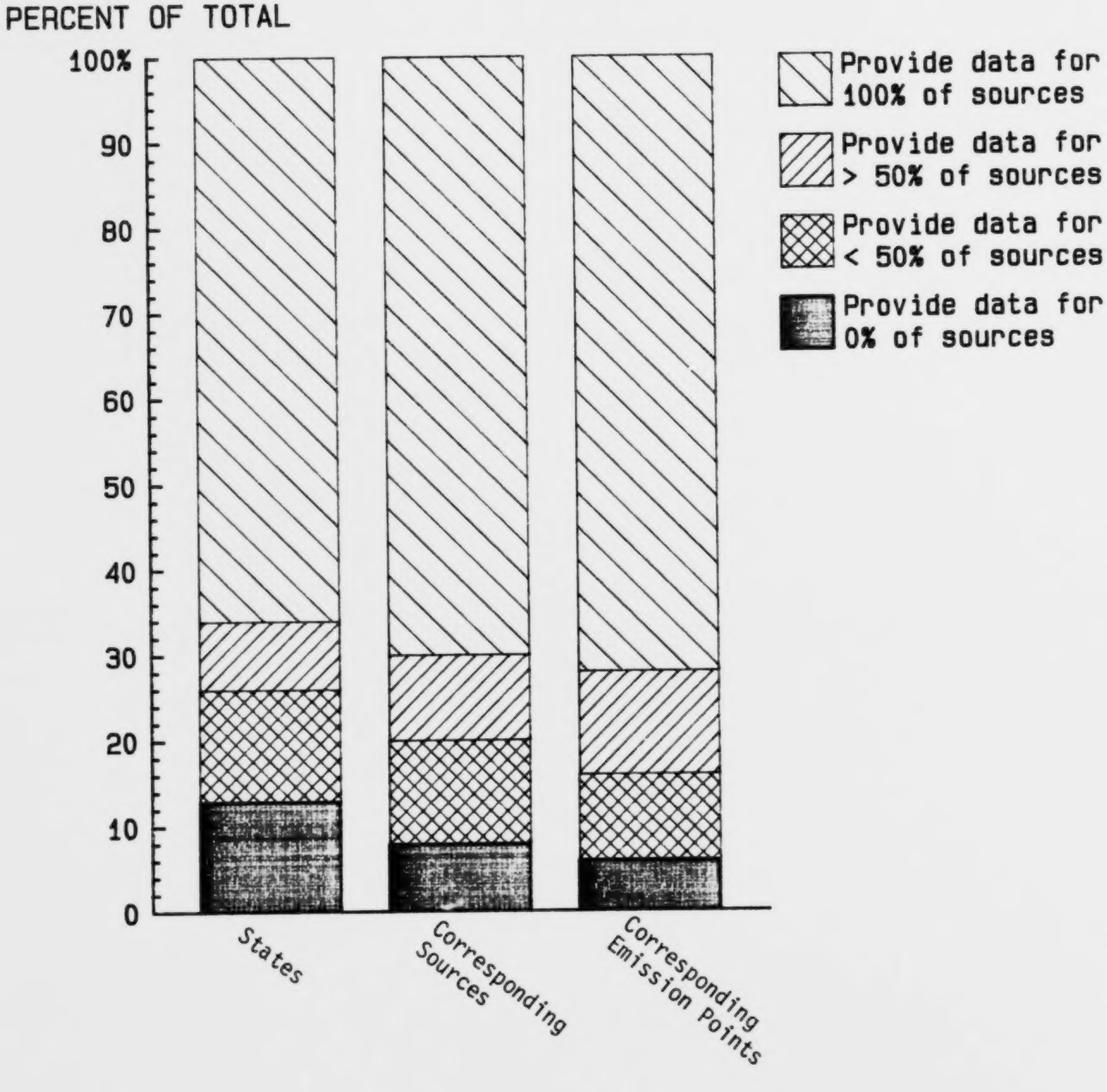


Figure 8. ABILITY TO PROVIDE 1985 EMISSIONS FOR NOx SOURCES > 2500 TPY (NAPAP Emissions Inventory)

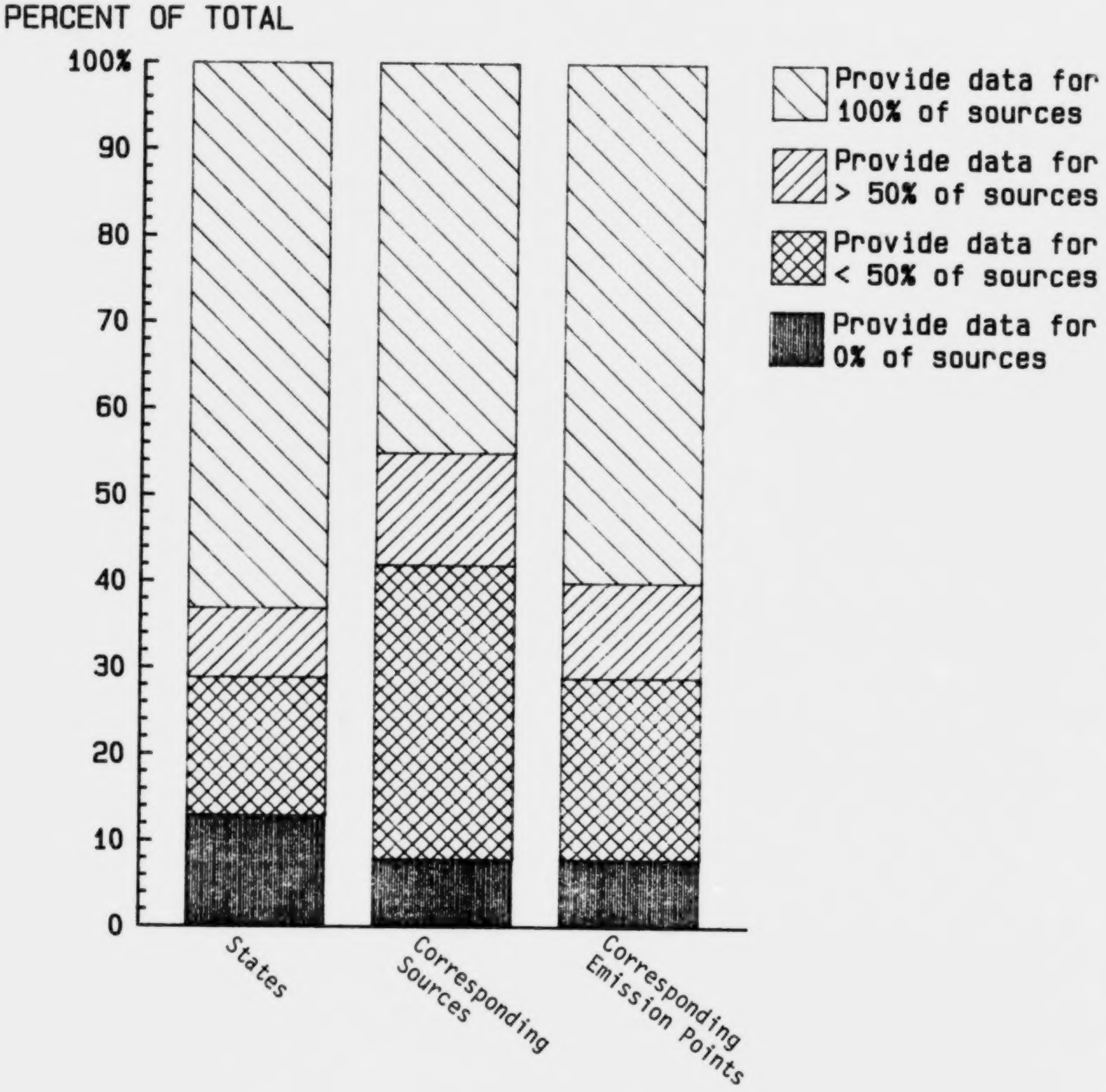
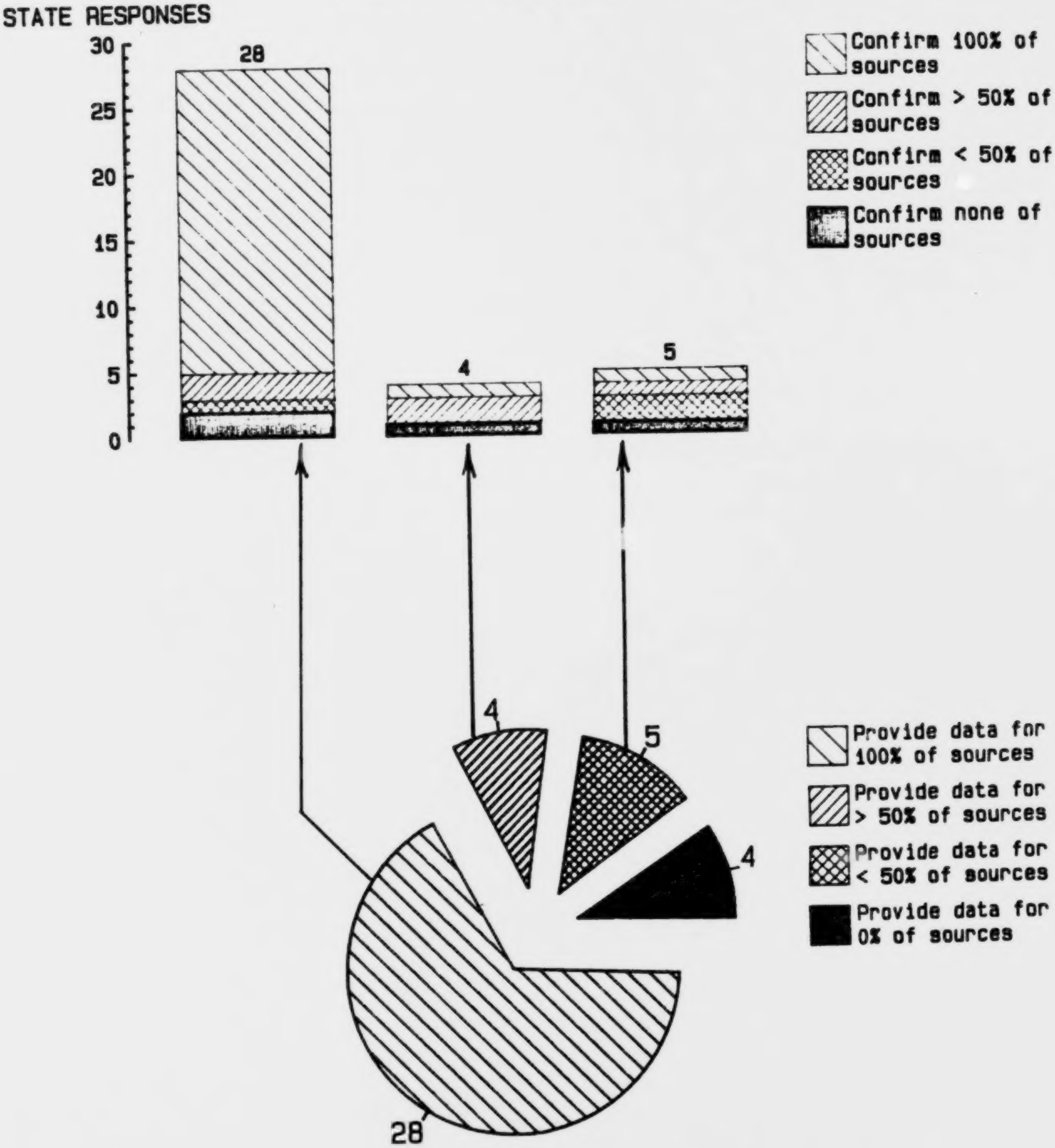


Figure 9. ABILITY TO CONFIRM 1985 EMISSIONS FOR SO2 SOURCES > 2500 TPY (NAPAP Emissions Inventory)



STATE CAPABILITIES TO PROVIDE DATA (Numbers of States)

Figure 10. ABILITY TO CONFIRM 1985 EMISSIONS FOR SO2 SOURCES > 1000 TPY (NAPAP Emissions Inventory)

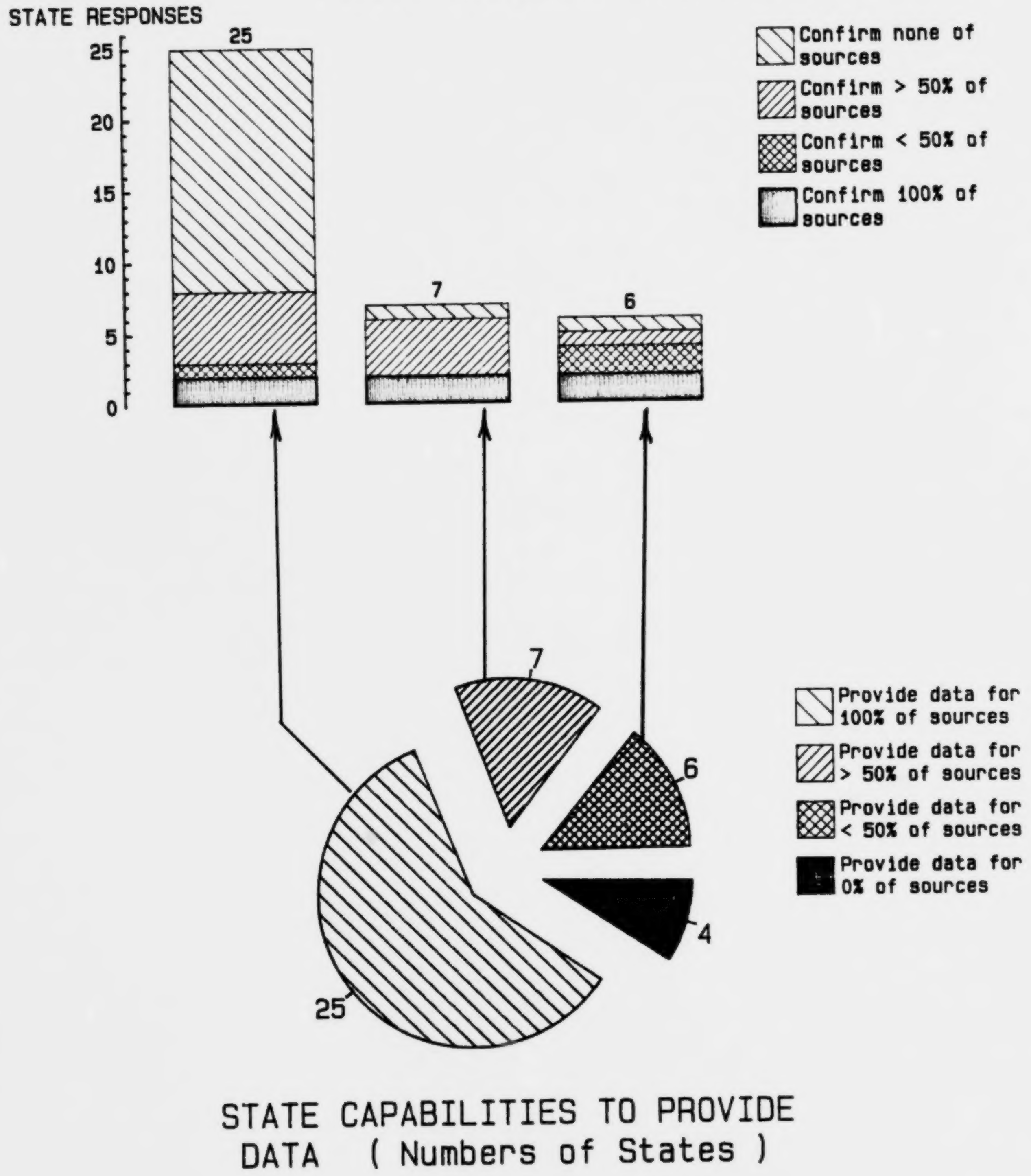
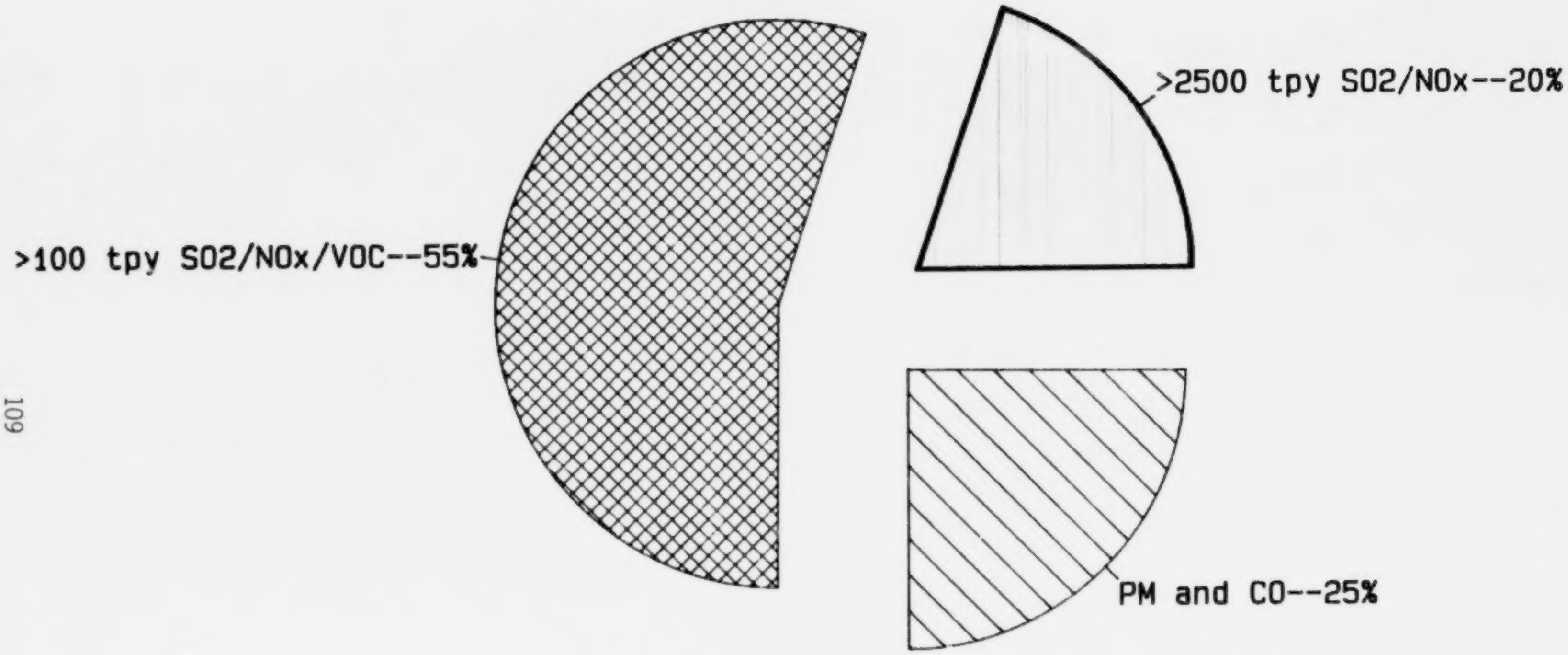


Figure 11. LEVEL OF ASSISTANCE REQUESTED TO DEVELOP 1985 NAPAP INVENTORY



Total Assistance Requested =
$ 2, 052, 688

An important indication from these numbers is that, based on the initial State calculations, available funds (about $1 million) could probably support the level of assistance needed to ensure the development of emission inventories of sources emitting more than 2,500 TPY of SO_2 or NO_x. However, the remaining funds do not appear sufficient to ensure adequate coverage of sources emitting more than 100 TPY of SO_2, NO_x, VOC, or PM or 1,000 TPY of CO. Subsequent evaluations will determine whether the assistance needed can be provided in a way to produce considerable cost savings, whether the data and inventory needs should be modified, or whether additional funding sources should be sought.

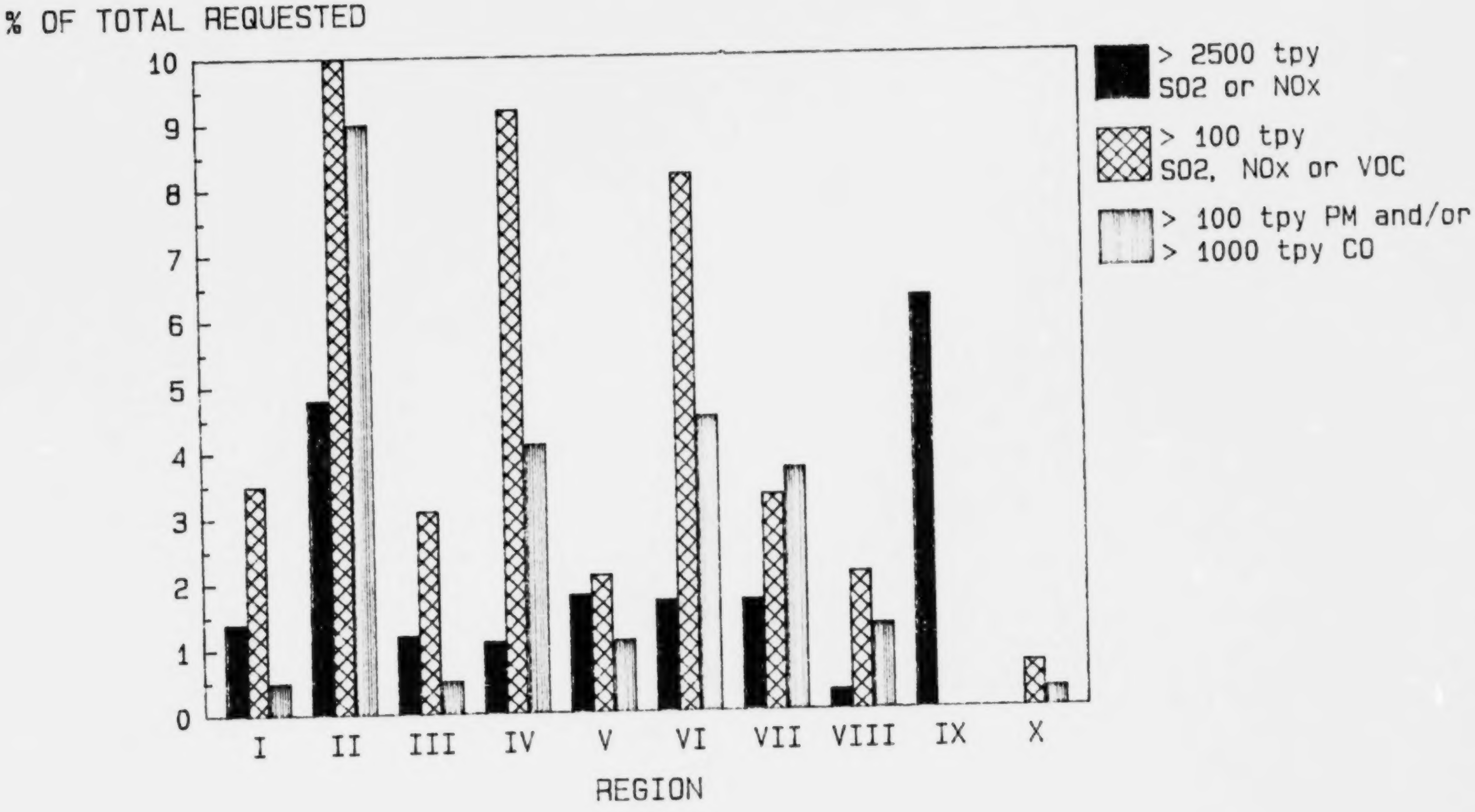
The same type of information on assistance requested has been summarized on a Regional basis. Figure 12 shows that acquiring emission data for sources emitting more than 100 TPY of SO_2, NO_x, or VOC accounts for the largest portion of requested assistance in almost all EPA Regions.

It is interesting to note that the 20 percent of requested assistance for developing emission data for the large SO_2 and NO_x sources would help ensure that over 90 percent of SO_2 point source emissions and over 80 percent of NO_x point source emissions are covered in the inventory effort. See Figure 13. On the other hand, the assistance requested for SO_2, NO_x, and VOC sources greater than 100 TPY would be associated with less than 10 percent of the SO_2 point source emissions and less than 20 percent of the NO_x emissions. It is important to add, however, that this later amount would also address about 95 percent of the VOC point source emission inventory.

As expected, the level of assistance needed generally increases with the number of sources a State has, although there are exceptions. Figure 14 illustrates the requested assistance for developing the inventory for sources emitting more than 2,500 TPY of SO_2 and NO_x as a function of the number of those sources in the State. Further evaluation reveals that the cost per source is widely variable and, if any trend can be discerned, it is generally constant over the range of the number of sources per State. Figure 15 shows the wide range for cost per source associated with the assistance requested for sources emitting more than 2,500 TPY SO_2 or NO_x. A similar graph for developing the inventory of sources emitting more than 100 TPY of SO_2, NO_x, VOC, or PM or 1,000 TPY of CO is shown in Figure 16. A slight downward trend is shown by the regression analysis, but the wide range in costs per sources is apparent.

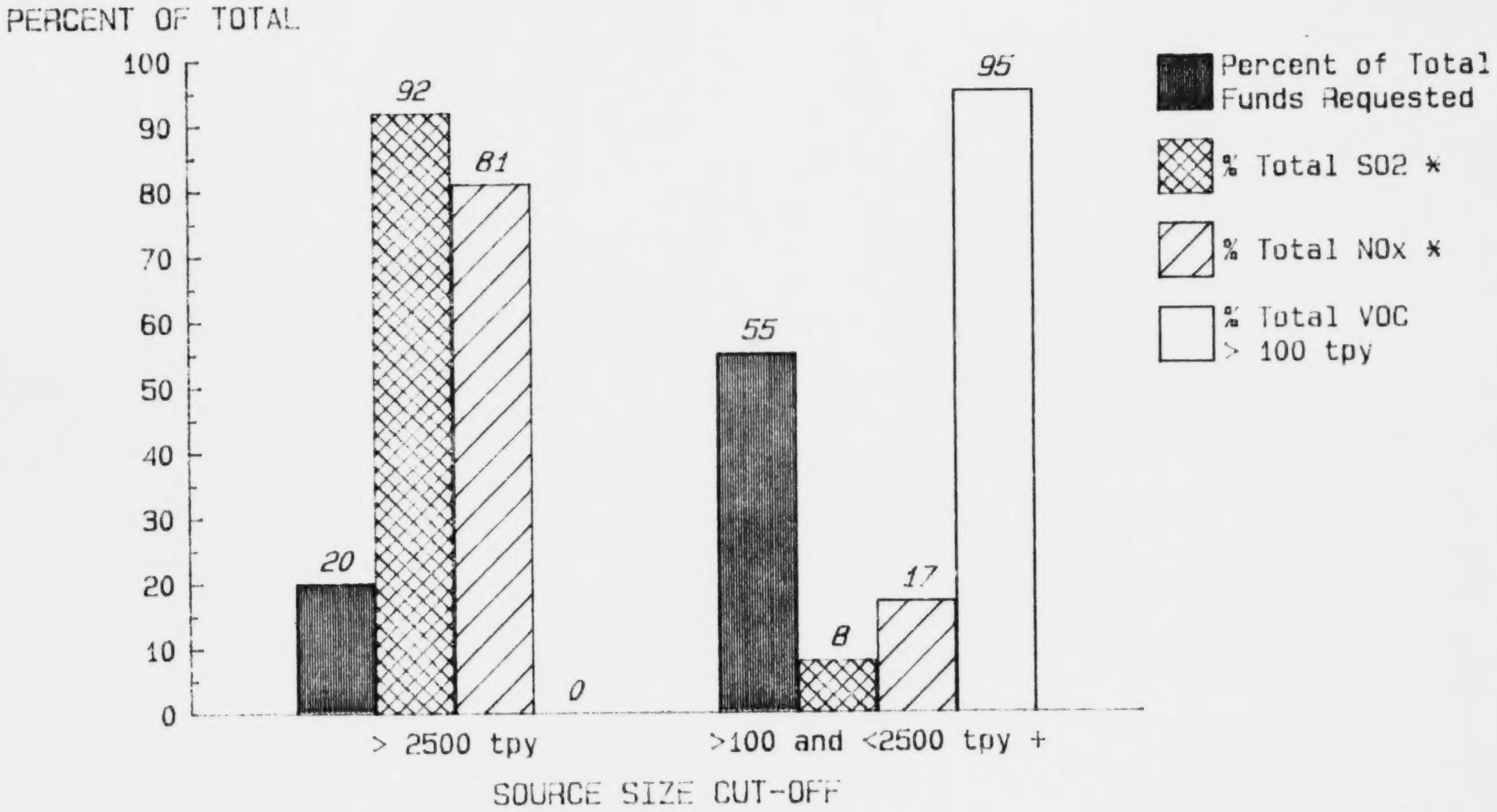
States indicated that most of the assistance needed would involve data editing and quality assurance activities, data collection from the sources, or data processing to enter the data into the State files. Figure 17 illustrate the responses of the States, given the seven categories listed. States also indicated a preference for receiving direct supplemental grant assistance as opposed to contractor support. See Figure 18.

Figure 12. LEVEL OF ASSISTANCE NEEDED FOR 1985 NAPAP INVENTORY EFFORT (by EPA Region)



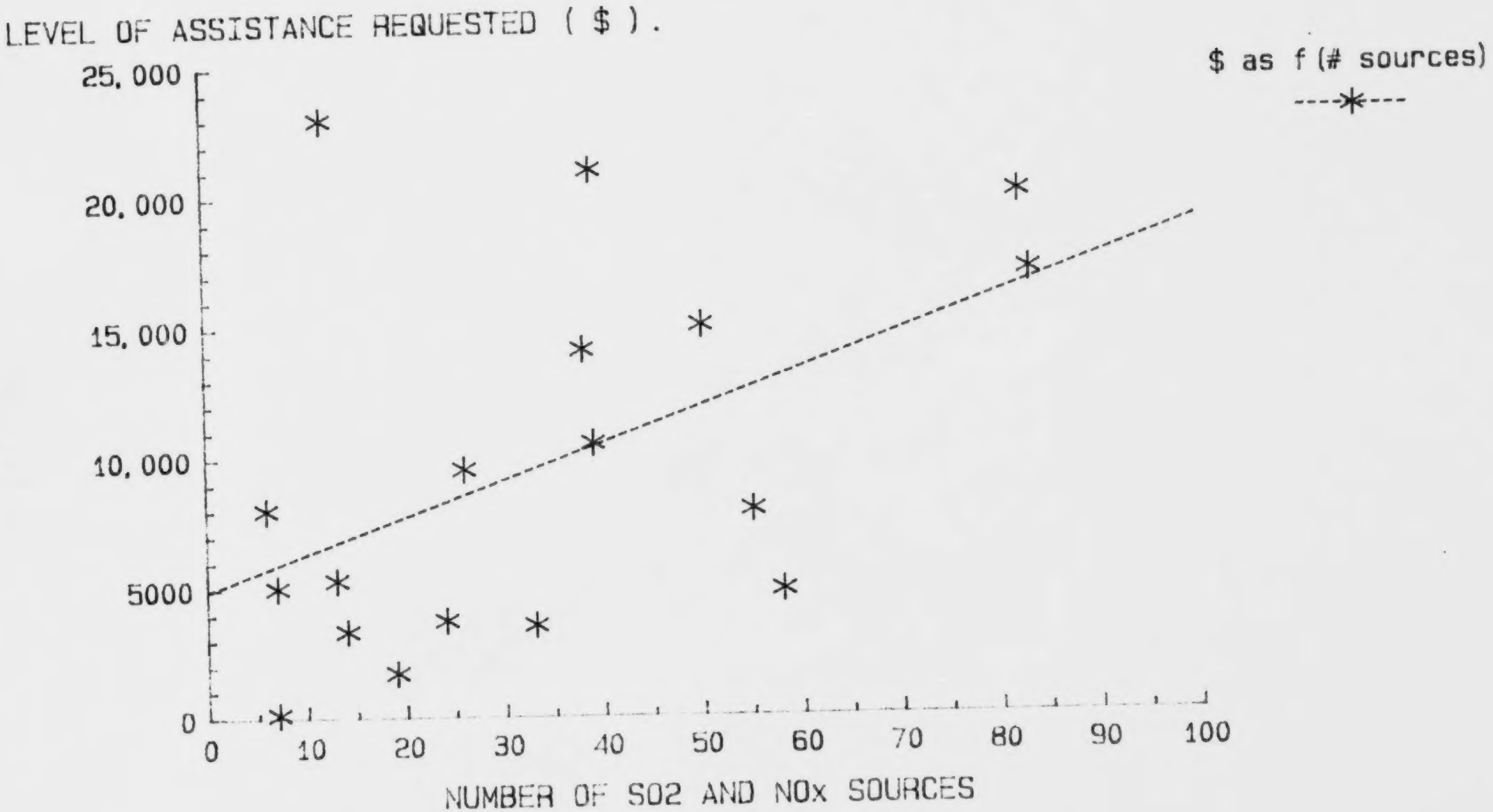
October 17, 1985

Figure 13. COVERAGE OF EMISSIONS INVENTORY AT TWO LEVELS OF ASSISTANCE



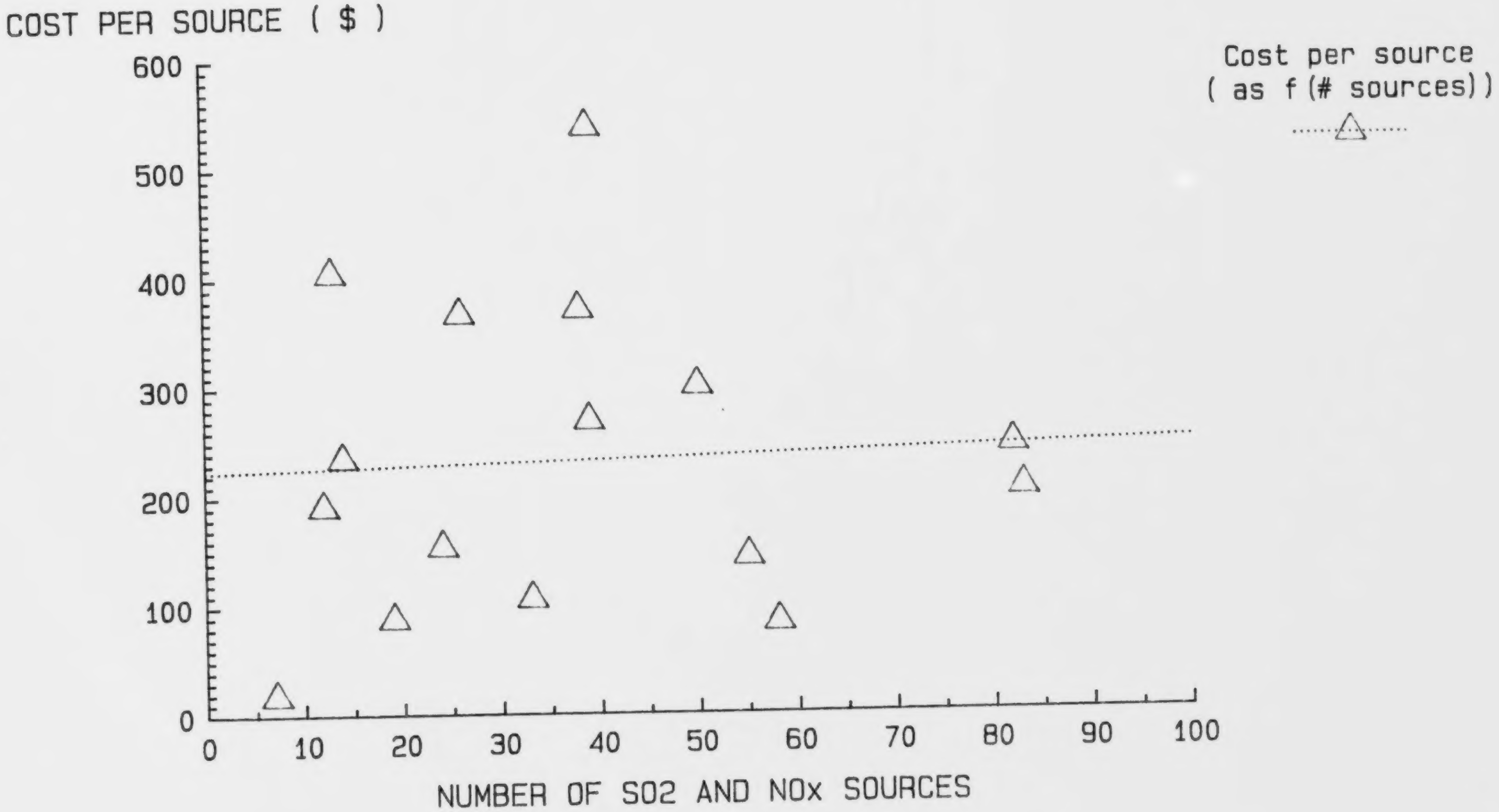
* Point sources only. (1980 NAPAP Inventory)
+ > 100 tpy VOC
October 16, 1985

Figure 14. NUMBER OF SO2 AND NOx SOURCES > 2500 TPY vs. LEVEL OF ASSISTANCE REQUESTED *



* Overlap may have occurred in adding SO2
and NOx sources.
October 10, 1985

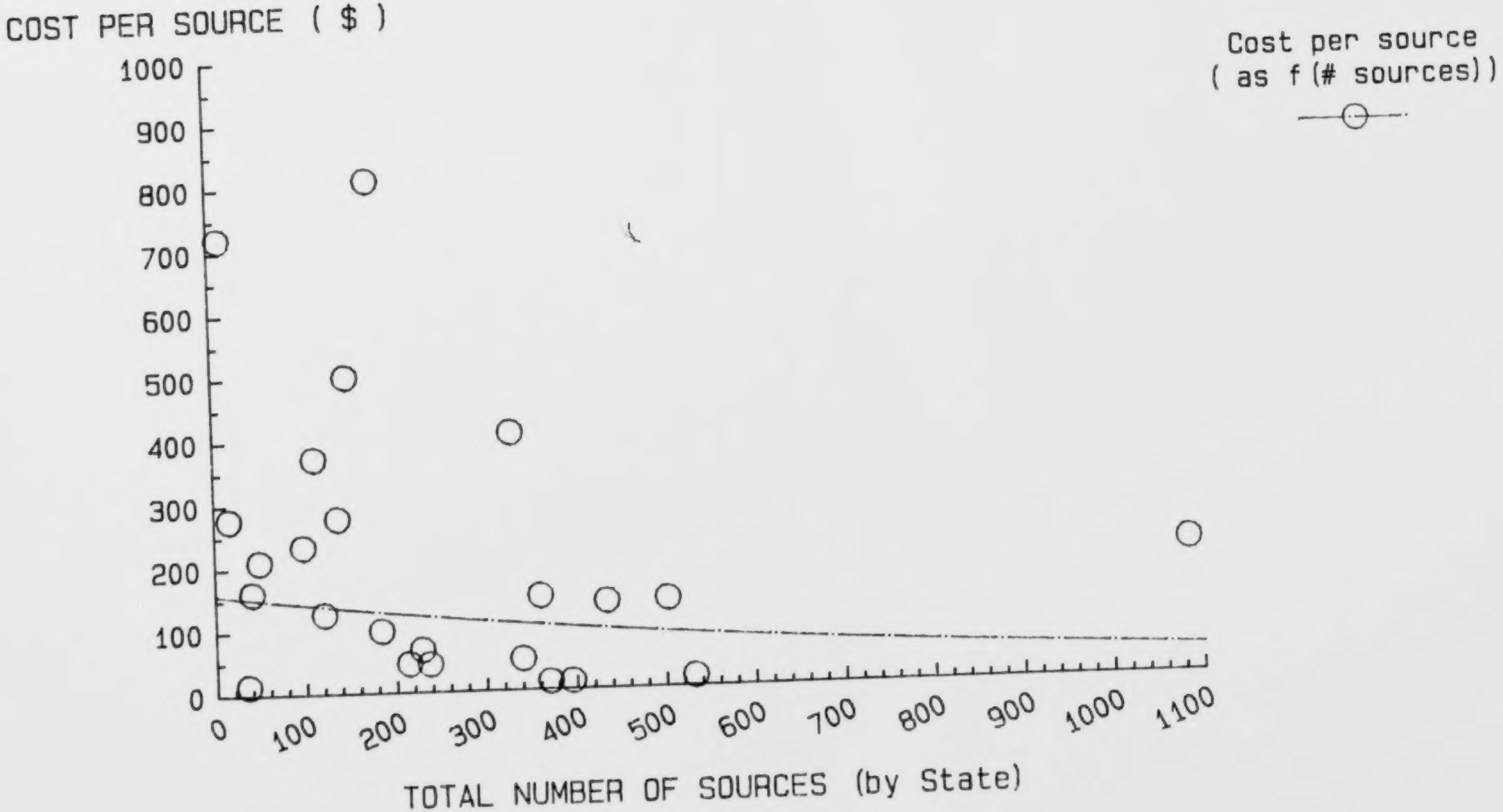
Figure 15. COST PER SOURCE (>2500 TPY SO2 or NOx) AS FUNCTION OF NUMBER OF SOURCES *



* Overlap may occur in adding SO2 and NOx sources.

October 10, 1985

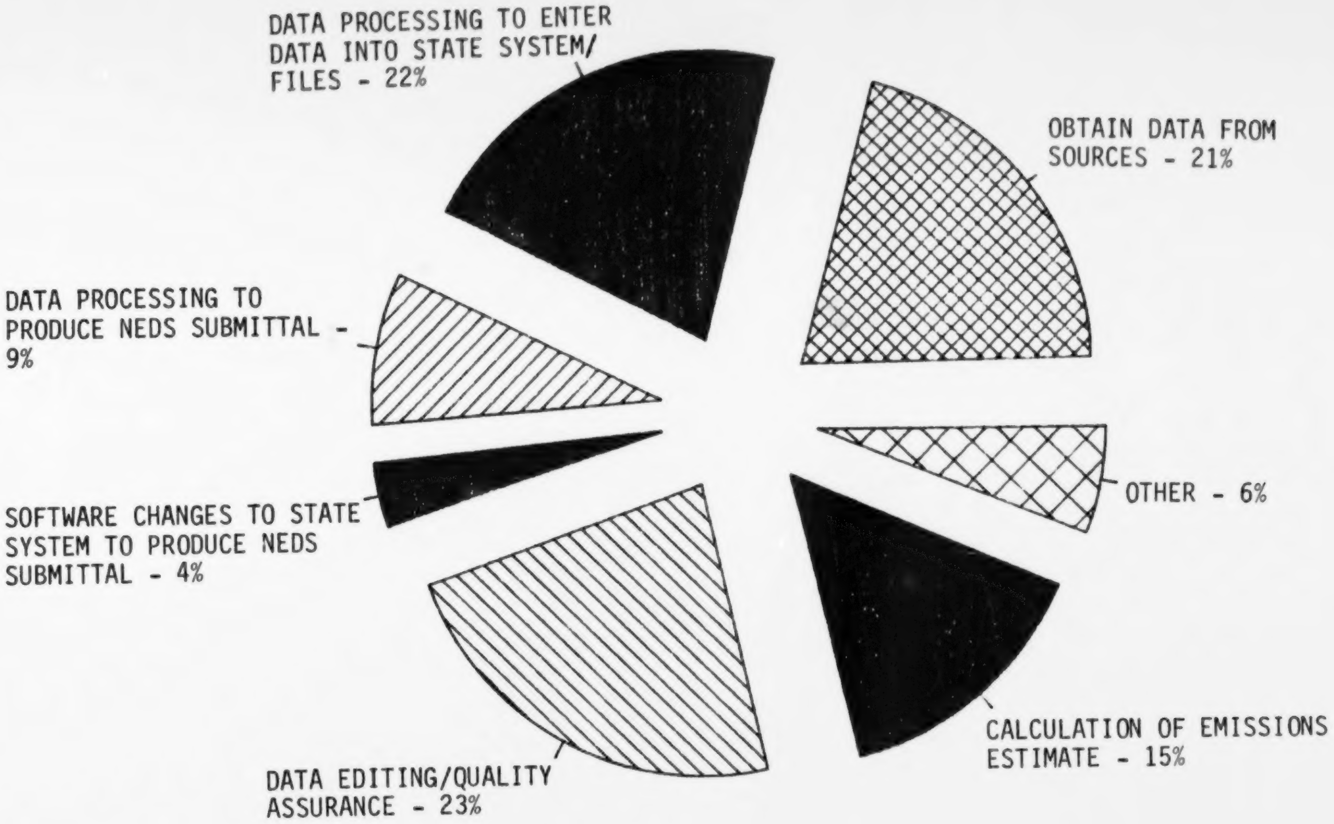
Figure 16. COST PER SOURCE, TOTAL INVENTORY (>100 tpy SO2, NOx, VOC and PM, and >1000 tpy CO)



* Overlap may occur in adding SO2 and NOx
sources.

October 10, 1985

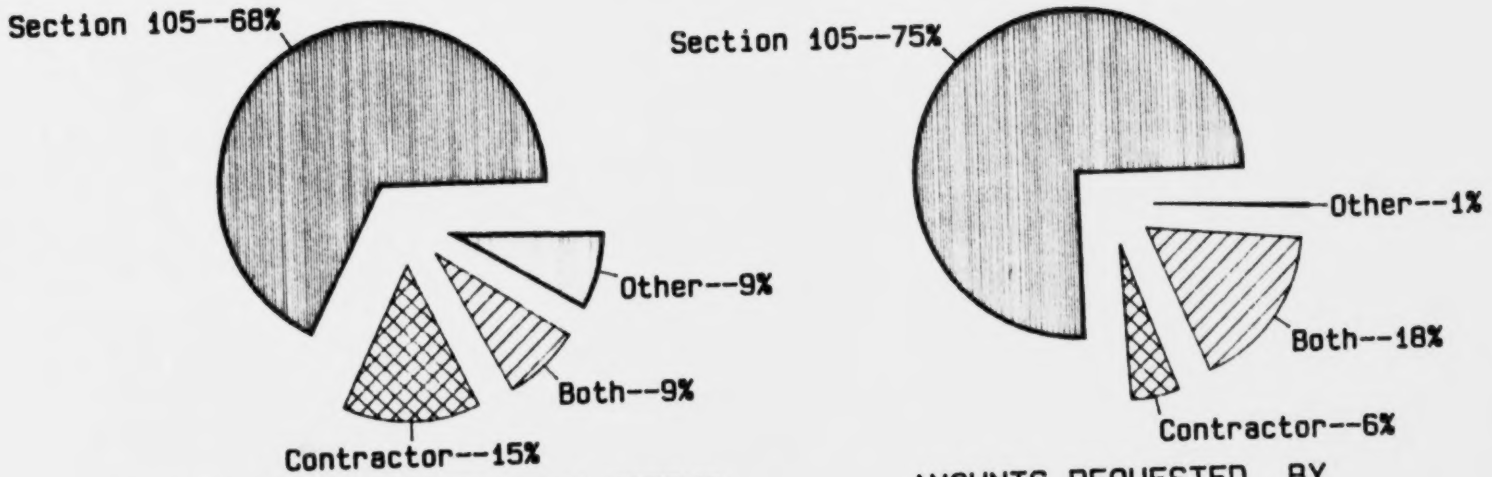
Figure 17. TYPES OF ASSISTANCE NEEDED
(1985 NAPAP INVENTORY)



42 of 49 total possible responses received.

October 17, 1985

Figure 18. ASSISTANCE REQUESTED BY STATES FOR 1985 NAPAP INVENTORY (by Type and Amount)



PROPORTION OF STATES REQUESTING ASSISTANCE, BY MECHANISM

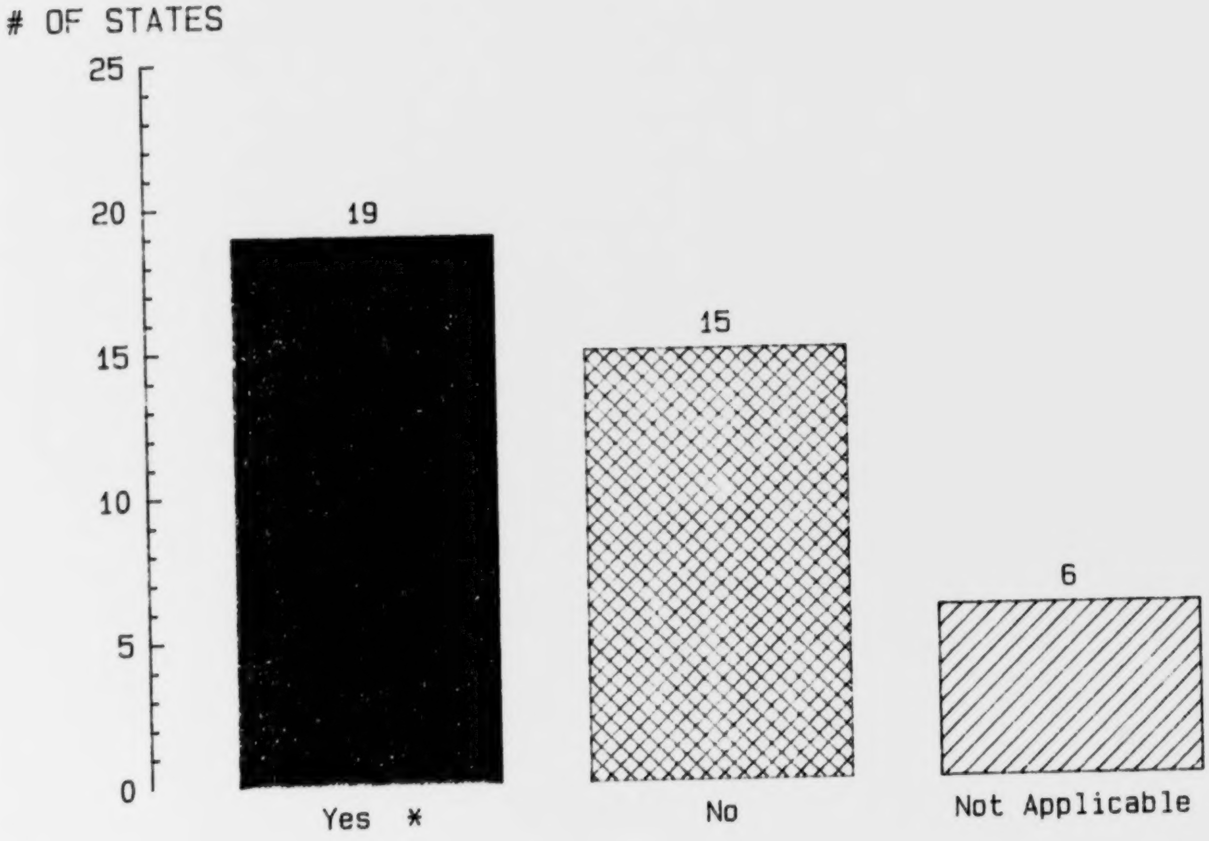
AMOUNTS REQUESTED, BY MECHANISM

Almost all States noted that they would not be able to submit the desired data significantly before July 1, 1987. Nineteen (19) States indicated that their requests for assistance would be reduced if two or three additional months were allowed for their submittal. See Figure 19. Eight of these 19 States were able to estimate a total reduction in resource requirements of about $325,000; the other States did not provide estimates of reductions.

Process for Providing Assistance

The survey responses will be used to determine the types and levels of assistance needed by States to provide the desired emission data. About $1 million ($500K in Section 105 funds and $500K in NAPAP funds) are available to provide direct financial assistance to States or to develop appropriate contractor projects to satisfy the needs of the States. In addition to the survey responses, follow-up discussion with States and Regional Offices will be used to identify alternative approaches for providing the assistance needed. The alternative approaches will be reviewed with the STAPPA committee, and a final plan for providing the assistance will be developed. By December 1985, appropriate grant amendments to States should be made and contractor projects should be developed.

Figure 19. WOULD ADDITIONAL TIME REDUCE RESOURCE REQUIREMENTS ?



* $325,000 identified as of October 16, 1985.

SESSION 3: FORMULATION OF MAN-MADE POLLUTANT EMISSION FACTORS

Chairman: Dale Pahl
Air and Energy Engineering Research Laboratory (MD-61)
U. S. Environmental Protection Agency
Research Triangle Park, NC 27711

DEVELOPMENT OF NAPAP EMISSION FACTORS FOR ANTHROPOGENIC SOURCES

BY

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EPA Contract No. 68-02-3994
Work Assignment No. 32

EPA Project Officer:
J. David Mobley
Air and Energy Enginering Research Laboratory
Research Triangle Park, NC

Presented At:

Second Annual Acid Deposition Emission Inventory Symposium

Charleston, SC

November 12-14, 1985

DEVELOPMENT OF NAPAP EMISSION FACTORS FOR ANTHROPOGENIC SOURCES

BY: J.B. Homolya
Radian Corporation

ABSTRACT

The NAPAP emissions inventory activities from 1980 to 1985 have included the development of anthropogenic emissions inventories to support the formulation and testing of an Eulerian atmospheric transport, transformation, and deposition model. The model requires high levels of species, temporal, spatial, and sectoral resolution for emissions inventory input parameters. A number of the pollutant species included in the inventory development required the formulation of source-categorized emission factors which were developed from assessments of data available in the literature. To date, detailed emission factor assessments have been prepared for primary sulfates, ammonia, hydrochloric acid, and hydrofluoric acid. For certain source categories, the assessments were compared with additional stack measurements conducted to address data gaps or to verify the quality of the emission factors. An assessment of alkaline dust emission factors has pointed to the need for both detailed chemical composition and particle size distribution data to support emission factor development. Ongoing research is addressing this need and providing further development of VOC emission factor data needed to support the 1985 NAPAP emissions inventory.

INTRODUCTION

The Eulerian acid deposition model currently being developed by the National Center for Atmospheric Research requires emissions data based on certain chemical species which act as direct acidic emissions to the atmosphere, scavengers of primary or secondary acids in the atmosphere, or catalysts in atmospheric transformation processes. Most of the chemical compounds or classes of compounds needed for input into the modules describing chemical transformation and deposition processes are non-criteria pollutants. Because of this, limited information is available concerning emission factors for most of these species. Therefore, an anthropogenic emission factor development program within NAPAP was designed to formulate appropriate emissions inventories using the best pollutant speciation data available. Emission factor development was initiated in 1983 with the preparation of emission factor assessments for primary sulfate[1], ammonia, hydrochloric acid, and hydrofluoric acid.[2] These species were selected because they are the priority pollutant species required for use in both Lagrangian modeling projects and the preliminary development of the Eulerian model.

The assessments began with a review of a compilation of data available in the literature. Where data were incomplete, a series of emission tests were conducted at the Department of Energy, Pittsburgh Energy Technology Center. To date, emissions tests have been conducted for bituminous coal combustion emissions of HCl, HF, and primary sulfate.

During 1985 preliminary assessments were performed on the development of point source alkaline particulate emission factors. Emphasis was given to the consideration of particle size distribution and the chemical composition of the alkaline fraction of total particulate emissions. Further, source categories were identified which require the development of VOC emission factors. Priorities have been assigned and incorporated in an implementation plan for data gathering.

This paper summarizes the current results of the NAPAP emission factor development program by presenting tabulated estimates of emission factors derived for the above species. The estimates are provided according to source classification code (SCC) format and include emissions calculations for 1980.

PRIMARY SULFATE EMISSIONS

Sulfur present in fossil fuels is emitted to the atmosphere mainly as SO_2. However, some of the sulfur is oxidized further in the combustion process and is emitted as primary sulfate compounds. The largest source of primary sulfate emissions (PSE) is from the combustion of coal and oil. The amount of primary sulfates emitted from a particular combustion source is dependent on a number of factors including fuel type and composition, combustion equipment design, operating parameters, and emissions controls.

A significantly higher conversion of fuel sulfur to primary sulfates occurs during oil firing as compared to coal firing. Enhanced PSE from oil combustion are due to the low ash, fast burning properties of fuel oil. Also, many residual oils contain high levels of vanadium which can catalyze the formation of SO_3 in a furnace.

Sulfates are formed in combustion processes in both the flame region and downstream in the heat transfer section. The major primary sulfate formation mechanisms include oxidation of SO_2 to SO_3, hydration of SO_3 to H_2SO_4, corrosion of boiler internals by H_2SO_4, and conversion of metallic oxides in fuel ash to particulate sulfates. In combustion flames, the predominant sulfate formation mechanism is reaction of molecular oxygen to form SO_3. The final SO_3 concentration leaving the flame zone depends on O_2 concentration, cooling rate, and the location and rate of mixing of excess air.

Increased PSE can be affected by the heterogeneous catalytic oxidation of SO_2 by metals, metal oxides, and soot suspended in the stream of combustion gases or deposited on boiler internal surfaces. Residual fuel oils from Venezuela and the Middle East contain significant amounts of vanadium which is liberated as V_2O_5 during combustion. V_2O_5 has been used extensively in the chemical manufacturing industry, as an oxidation catalyst. The major constituents of coal fly ash, SiO_2 and Al_2O_3, are only weak catalysts.

Sulfuric acid is formed by reaction of SO_3 with water vapor in combustion product gases. The acid can adsorb on ash or soot particles, condense on cooler parts of the combustion equipment, or be emitted to the atmosphere as a mist. The SO_3-to-H_2SO_4 conversion is temperature- and moisture-dependent.

REVIEW OF AVAILABLE PRIMARY SULFATE EMISSION FACTOR DATA BASES

Prior to the current need for NAPAP Task Group B to inventory PSE, two separate environmental program activities have included assessments of PSE factors for use in emissions inventory development. The Electric Power Research Institute sponsored a sulfate regional experimental study which included a summary of existing measurements data on primary sulfates and recommended generalized emission factors for a number of source categories. Many of the factors were based upon an extrapolation of a limited data set for uncontrolled fossil fuel combustion sources along with data contained in the EPA Emission Factor Guidelines (AP-42).

The United States/Canada Work Group 3B (WG 3B) prepared an emission inventory report in accordance with the Memorandum of Intent on Transboundary Air Pollution of August 1980. The report included a study[3] of U.S. and Canadian PSE using factors abstracted from the literature along with unpublished emissions data from Canadian measurements obtained from the use of a variety of sampling and analysis methods.

Following review of the EPRI and WG 3B sulfate emissions data sets, an extensive literature review was conducted to include all contemporary PSE measurements data in a final analysis and tabulation of emission factors appropriate for the NAPAP emissions inventory. Where available, all field measurements using the controlled condensation system (CCS) procedure were considered as the prime data set. Emissions data acquired through the use of methods other than CCS were included only if multiple measurements yielded precise data. Sulfate emission assessments were aggregated for different point sources within the same source category only if fuel composition and emission controls were similar.

PSE factors recommended for use in the NAPAP Emissions Inventories are listed in Table 1. The factors are reported for source classification codes (SCCs) and reflect sulfate emitted as SO_4 (molecular weight = 96) based on the chemical analyses of source test samples. Much of the current data set is for fossil fuel combustion in the industrial and utility sectors which contribute most of the regional mass emissions of primary sulfates. Table 1 also contains an estimate of the uncertainty for each emission factor. The estimates are based on a qualitative assessment of the representation of available source test

TABLE 1. PRIMARY SULFATE EMISSION FACTORS FOR NAPAP EMISSIONS INVENTORY

Source category	NEDS Source classification code (SCC)	Control device	Primary sulfate## emission factor	Uncertainty range*
Electric Utilities - External Combustion				
Eastern bituminous coal	1-01-002	ESP	0.385 lb/ton	A
		ESP and FGD	0.250†	C
Western bituminous coal	1-01-002	ESP	1.290	B
		ESP and FGD	0.761	C
Lignite	1-01-003	ESP	1.951	C
Residual oil (>1% sulfur content)	1-01-004	Fuel oil additive	5.439 lb/1,000 gallons	B
Industrial - External Combustion				
Eastern bituminous coal	1-02-002	Multiclones	2.646 lb/ton	B
		Multiclones and FGD	0.462	C
Residual oil	1-02-004	Multiclones	5.296 lb/1,000 gallons	D
		Multiclones and FGD	2.616	D
Commercial/Institutional - External Combustion				
Residual oil (<1% sulfur content)	1-03-004	Fuel oil additive	25.07 lb/1,000§ gallons	C
Space Heating - External Combustion				
Distillate oil	1-05-001-05	None	5.65 lb/1,000 gallons	C
Industrial Process - Chemical Manufacturing				
H_2SO_4-contact process	3-01-023	Demister	0.100 lb/ton acid produced	B
Industrial Process - Primary Metals				
Primary copper smelters	3-03-005-1	ESP	22.5 lb/ton concentrated ore	C
	3-03-005-2	ESP	1.08	C
	3-03-005-3	ESP	5.76	D
	3-03-005-4	ESP	15.66	D

(continued)

TABLE 1. Concluded

Source category	NEDS Source classification code (SCC)	Control device	Primary sulfate## emission factor	Uncertainty range*
Industrial Process - Primary Metals (Cont'd)				
Primary zinc smelters	3-03-030	ESP	55.5 lb/ton processed	D
Primary aluminum smelter	3-04-001	ESP	0.5% of SO_2	D
Iron production	3-03-008	ESP	2.0% of SO_2	D
Coke	3-03-003	Baghouse	0.320 lb/ton coal charged	D
Industrial Process - Wood Products				
Kraft pulp mill	3-07-001	ESP	#	C
Sulfite pulp mill	3-07-002	ESP	**	C
Wood/bark waste	1-02-009	Multiclones	3.6 lb/ton bark	D
Industrial Process - Mineral Products				
Cement manufacturing	3-05-006 and 3-05-007	ESP	††	D
Gypsum manufacturing	3-05-015	ESP	§§	D
Industrial Process - Petroleum Industry				
Fluid crackers	3-06-002	ESP	15.0 lb/1,000 barrels oil	C
Sulfur recovery Claus plants	3-01-032	None	2.8 lb/ton produced	C

* Estimated emission factor uncertainty. Assumes that 90% of the values for an individual source lie within the mean uncertainty estimates. Corresponding values are: A = ±10%; B = ±25%; C = ±50%; and D = ±75%.

† Emission factor based on average sulfate scrubbing efficiency of 35%.

§ Emission factor applicable only to low sulfur content (0.3% S) residual fuel oil.

# Total sulfate emissions for kraft pulp mills estimated as 85% of NEDS total particulate emissions from kraft recovery boilers.

** Total sulfate emissions from sodium-base sulfite mills estimated as 70% of NEDS SO_2 emissions; for calcium-base sulfite mills estimated as 25% of NEDS SO_2 emissions.

†† Total sulfate emissions from cement kilns estimated as 5.6 lb/ton of cement on an uncontrolled basis. Average particulate control efficiency from NEDS data assumed to apply in order to calculate actual emissions.

§§ Total sulfate from gypsum plants estimated as 56% as NEDS actual particulate emissions.

## Although it is EPA policy to utilize metric units, non-metric units have been used in this paper since the NAPAP emission factor file is in non-metric units. For conversion from non-metric units to metric units note, that 1 lb = 0.453 kg and that 1 ton (non-metric) = 1 short ton = 2000 lbs = 0.907 metric tons.

data to the assignment of SCC-level factors. Improvements are needed in the data base for low sulfur residual-oil-fired industrial and commercial boilers which represent a significant source of sulfur emission in major metropolitan areas in the eastern U.S. Also, sulfate emission from the pulp and paper industry needs further refinement. Pulp mill operations are concentrated in the acid deposition sensitive Northeast and represent a major SO_2 and particulate source contributor in the southeastern United States.

HYDROCHLORIC ACID EMISSIONS

Table 2 lists the sources of hydrochloric acid emissions along with emission factors and estimates of emissions for 1980. Coal combustion is the largest source of HCl emissions, accounting for 467,000 ton/yr.

Coal is formed over eons from successive layers of fallen vegetation. As vegetation accumulates, physical and chemical changes, such as loss of water and volatile matter, occur. Over time, the vegetation turns from peat to lignite, the earliest stage in the formation of coal. As lignite is compressed with deeper burial, the heat associated with the compression drives off volatile components. As more volatile components are driven off, the rank and quality of the coal increase from lignite to subbituminous, bituminous, and anthracite. The chlorine concentrations of domestic coals vary over a large range. The coal fields with the highest chlorine content are in the Illinois basin. The molecular form of chlorine in coal is not clearly understood. Chlorine in coal exists in both organic and inorganic forms. Although it has been shown that chlorine occurs in coal in a form more volatile than sodium chloride, no correlation has been developed between the molecular form of chlorine in coal and its combustion products.

COAL COMBUSTION IN UTILITY BOILERS

There are three principal types of coal-fired utility boilers: stoker-fired, cyclone furnaces, and pulverized-coal-fired (PC-fired). Of the three types currently used, PC-fired is the most common. Scrubbers, electrostatic precipitators (ESP), cyclones, and baghouses

TABLE 2. MAJOR SOURCES OF HYDROCHLORIC ACID EMISSIONS

Source	Emission factor* (lb emitted/unit)	1980 Emissions (ton/yr)	SCC	SCC units
Coal combustion		467,000		
Utility and industrial				
bituminous	1.9		1-01, 02-002	ton burned
lignite	0.01		1-01, 02-003	ton burned
Commercial/institutional				
anthracite	3.07		1-03-001	ton burned
bituminous	1.48		1-03-002	ton burned
lignite	0.351		1-03-003	ton burned
Incineration		75,000		
Municipal refuse				
multiple chamber	5.0		5-01-001-01	ton burned
single chamber	5.0		5-01-001-02	ton burned
conical design-refuse	5.0		5-01-005-07	ton burned
Industrial wastes				
multiple chamber	5.35		5-03-001-01	ton burned
single chamber	5.35		5-03-001-02	ton burned
controlled air	5.35		5-03-001-03	ton burned
conical design-refuse	5.35		5-03-001-04	ton burned
Liquid wastes	1.19		NA	ton burned

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are used frequently as flue gas control techniques. These control devices are effective for different pollutants and often are used in combination. An ESP or a baghouse has no demonstrated effect on HCl emissions. However, alkaline materials are sometimes used in conjunction with a baghouse to remove sulfur compounds. This could also result in a removal of HCl.

Cyclones or centrifugal separators also are used in the control of particles from utility boilers. Cyclones are assumed to have no effect on HCl emissions.

Wet scrubbers also are used to remove particles from flue gas streams. A wet scrubber can use a variety of methods to wet the contaminant particles in order to remove them from the gas stream. The efficiency of a wet scrubbing device for HCl emissions control should be greater than that of a baghouse or ESP. One study demonstrated an 80 percent efficiency for HCl removal from bituminous-coal-fired utility boilers.

Gaseous sulfur compounds typically are removed from flue gas streams by flue gas desulfurization (FGD) systems. In FGD, the sulfur compounds are reacted with an alkaline substance. The resulting compound then is removed from the flue gas stream. HCl should be removed from flue gases with FGD systems; however, little data are available to quantify the removal efficiency.

COAL COMBUSTION IN INDUSTRIAL BOILERS

Industrial boilers are smaller than utility boilers and are used by industry to provide steam or hot water. Only 10 to 15 percent of the industrial boiler coal consumption is used for electricity generation. Coal burning industrial boilers are classified by heat transfer method, arrangement of heat transfer surface, and fuel feed system. The three types of heat transfer systems are watertube, firetube, and cast iron. Most newly installed industrial boilers are either watertube or firetube. Fuel feed systems are the same as discussed under utility boilers--stoker, cyclone, and PC-fired. Emission factors for HCl have not been developed for industrial boilers. Emission factors developed for utility boilers can be used for industrial boilers when the type of boiler and fuel are the same. Air pollution control technologies for industrial boilers are the same as those discussed under utility boilers.

COAL COMBUSTION IN RESIDENTIAL BOILERS

At the present time, approximately 1 percent of the nation's homes are heated with coal. Lignite is burned for residential heating in North Dakota only; anthracite combustion is limited to northern states east of the Mississippi River, with 64 percent in Pennsylvania; and bituminous coal is burned in every state except Connecticut, Delaware, Maine, Maryland, New Hampshire, New Jersey, North Dakota, Rhode Island, and Vermont. HCl emission factors have been reported as 60.5×10^{-9}, 120.0×10^{-9}, and 35.1×10^{-9} lb/Btu for bituminous, anthracite, and lignite combustion, respectively.

INCINERATION

HCl is emitted during the incineration of wastes containing chlorinated hydrocarbons, chlorine-containing plastics such as polyvinyl chloride, and chlorine or chlorides, often in the form of common salt. Incineration is a combustion process used to change the chemical or physical characteristics of waste by oxidation which, even under ideal combustion conditions, results in inorganic solid residues and exhaust gases.

HCl is emitted as a result of incomplete combustion from three types of incinerators: (1) municipal incinerators that burn public garbage; (2) municipally owned incinerators that burn primarily industrial wastes; and (3) incinerators that burn liquid wastes such as polychlorinated waste, waste oil, and various other hydrocarbon liquids. Emissions from the public incineration of approximately 30×10^{5} tons of collected refuse in the United States in 1970 produced estimated emissions of 75,000 tons of HCl. The emission factor for this source is calculated as 5 lb HCl/ton of refuse burned.

Emission factors for municipally owned industrial waste incinerators range from 2.0 to 7.0 lb HCl/ton of wastes burned with a mean value of 5.35 lb/ton. Several sources were tested for emissions from incineration of liquid wastes. Emission factors from the test data ranged from 0.72 to 1.61 lb HCl/ton of waste burned with a mean uncontrolled emission factor of 1.19 lb/ton.

HYDROFLUORIC ACID EMISSIONS

Table 3 shows the major HF emissions sources and provides a summary of total emissions, emission factors, and applicable SCCs. Coal combustion is the largest source of HF emissions, accounting for 58,000 tons/yr. Fluorine present in coal is converted to HF during the combustion process and emitted to the atmosphere. The effectiveness of emissions controls for HF is assumed to be similar to that discussed previously for HCl.

PRIMARY ALUMINUM INDUSTRY

The base ore for primary aluminum production is bauxite, a hydrated oxide of aluminum consisting of 30 to 70 percent alumina (Al_2O_3) and lesser amounts of iron, silicon, and titanium. Bauxite ore is first purified to alumina by the Bayer process, and then the alumina is reduced to elemental aluminum. The production of alumina and the reduction of alumina to aluminum are two separate processes, seldom accomplished at the same location. Fluoride emissions from primary aluminum production are from the reduction process only.

Fluoride emissions from primary aluminum production occur in the gaseous phase as HF with a small amount of silicon tetrafluoride (SiF_4), and in the particulate phase as cryolite (Na_3AlF_6). The ratio of gaseous to particulate fluorides varies with the particular emission point within the process.

The principal points of HF emissions are the primary and secondary emissions from the potrooms housing the reduction cells and, in the case of the prebake cell, the emissions from the associated anode bake plants. HF emissions from bauxite grinding and aluminum hydroxide production are negligible. The manufacture of anodes for prebake cells emits HF from the fluorides in the recycled anode butts.

TABLE 3. MAJOR SOURCES OF HYDROFLUORIC ACID EMISSIONS

Source	Emission factor* (lb emitted/unit)	1980 Emissions (ton/yr)	SCC	SCC units
Coal combustion		58,000		
Utility				
bituminous	0.23		1-01-002	ton burned
lignite	0.01		1-01-003	ton burned
Industrial				
bituminous	0.62-0.81		1-02-002	ton burned
Commercial/institutional				
anthracite	0.13		1-03-001	ton burned
bituminous	0.17		1-03-002	ton burned
lignite	0.063		1-03-003	ton burned
Primary aluminum production		13,300		
Anode baking furnace	0.52		3-03-001-05	ton produced
Prebaked reduction cell	4.9		3-03-001-01	ton produced
Prebaked fugitive emissions	1.2		3-03-001-08	ton produced
Vertical Soderberg stud cells	0.6		3-03-001-03	ton produced
Vertical Soderberg stud cells fugitive emissions	4.9		3-03-001-10	ton produced
Horizontal Soderberg stud cells	1.9		3-03-001-02	ton produced
Horizontal Soderberg stud cells fugitive emissions	2.2		3-03-001-09	ton produced
Gypsum ponds	1.275	6,400	3-01-016-02	tons P_2O_5 produced

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All three types of reduction cells emit HF from thermal hydrolysis of volatilized bath materials, particularly cryolite. The reaction of solid and vaporized fluorides at elevated temperatures occurs primarily at the point where the hot gases escape through vents in the crust at the cell surface. The hydrogen required for the formation of HF is supplied in part from water vapor in the air. Other sources of hydrogen include residual moisture in the alumina and bath raw materials and hydrocarbons in the carbon anodes. The HF emission factor for prebake cells is the sum of the HF emission factors for the anode baking furnace and the prebake reduction cell.

AMMONIA EMISSIONS

Most ammonia emissions result from natural and biological processes, primarily through the decomposition of organic matter such as dead plants and animal and human excrement. Other natural sources include forest fires and volatilization from land and ocean masses. Table 4 lists the major sources of ammonia emissions. Emission factors are given along with the total estimated emissions.

AMMONIA FROM LIVESTOCK WASTES

The major sources of ammonia emissions from the production of livestock include beef production operations and manure spreading. Production of beef cattle can be divided into four stages: calf production, backgrounding, finish feeding, and slaughtering. The wastes excreted by cattle grazing on pastures or range areas are disperse, allowing the soil to assimilate the excrement. However, manure produced in feedlot situations must be removed and processed because of space restrictions. Animal manure contains complex carbohydrates, protein, and fats which are metabolized into simpler compounds by microorganisms. In the presence of oxygen, the end products of metabolism are heat, CO_2, and H_2O. This process, called aerobic metabolism, is dependent upon temperature, oxygen, and moisture. In the absence of oxygen, anaerobic metabolism occurs with ammonia as an end product.

TABLE 4. MAJOR SOURCES OF AMMONIA EMISSIONS

Source	Emission factor* (lb emitted/unit)	1980 Emissions (ton/yr)	SCC	SCC units
Livestock wastes				
Manure field application				
beef cattle	34	1,900,000	NA	per animal
dairy cows	71	390,000	NA	per animal
hogs	7.5	240,000	NA	per animal
broilers	0.47	920,000	NA	per animal
other chickens	0.47	56,000	NA	per animal
Beef cattle feedlots	2.1	24,000	3-02-020-02	per animal
Coal combustion	2.0	525,000	1-02, 02, 03	ton burned
Ammonium nitrate manufacture				
Neutralizer	0.86-36.0	3,100-130,000	3-01-027	ton produced
Solids formation	0.54-33.4	920- 57,000	3-01-027	ton produced

*Although it is EPA policy to utilize metric units, non-metric units have been used in this paper since the NAPAP emission factor file is in non-metric units. For conversion from non-metric units to metric units, note that 1 lb = 0.453 kg and that 1 ton (non-metric) = 1 short ton = 2000 lbs = 0.907 metric tons.

Ammonia is emitted from beef production operations as a result of anaerobic decomposition and volatilization of cattle wastes. Anaerobic decomposition occurs in the solid manure beneath the feedlot surface, in manure stockpiles, and in runoff holding ponds. Feedlot disturbances, such as mounding and manure removal, greatly increase the release of ammoniacal compounds to the atmosphere. Although 25 to 40 percent moisture levels are necessary on feedlot surfaces for aerobic decomposition, too much rain or watering, resulting in puddling and wet spots, can trigger increased ammonia production. In addition, about half of the nitrogen that cattle excrete is in the form of urea in cattle urine. The urea in urine is hydrolized to ammonia and can be released to the atmosphere.

The spreading of livestock manure on agricultural land is another source of ammonia emissions. Field experiments indicate that, when dairy manure was spread in the field preparation for plowing under, an 85 percent loss of total ammonia by volatilization occurred. The dairy manure volatilization rate may be applied to other types of domestic livestock manure. The ammonia emission estimates from the field application of livestock manure given in Table 4 were derived by multiplying ammonia excretion rates by animal population levels for 1980 and by the 85 percent loss factor. Manure from beef cattle is the major source of these emissions. The total emissions estimate for ammonia from field application of livestock waste is 3.5 million tons.

AMMONIA NITRATE MANUFACTURE

Ammonia nitrate is produced by mixing nitric acid and ammonia in a reactor or neutralizer. Either ammonia or nitric acid is emitted from the neutralizing depending upon which reactant is added in excess to the process. Most plants today operate the neutralizer with excess ammonia. An 83 percent aqueous ammonium nitrate solution is produced in the neutralizer and may be sold for use as a fertilizer or further concentrated for use in solid ammonium nitrate formation.

COAL COMBUSTION

Ammonia emissions from coal combustion were estimated from a limited set of measurements data. An emission factor of 2 lb NH_3/ton coal burned was applied to all external combustion boilers for anthracite, bituminous, and lignite fuels.

ALKALINE DUST EMISSION FACTORS

Anthropogenic point source alkaline particulate emission factors have been calculated from available alkaline dust emissions data reported by Meteorological and Environmental Planning, Ltd. and the Ontario Research Foundation.[4] The emissions data were based on the identification of major anthropogenic sources of particulate Ca, Mg, Na, and K emissions. Total particulate emissions from these sources were estimated along with the amount of the alkali metals contained in the particulate.

Table 5 is a summary of emission factors for each alkali metal assigned to individual SCC categories. The basic assumption is that the alkali metal contents of U.S. and Canadian particulate emissions are identical. For power generation and fuelwood combustion, the emission factors are assumed to be applicable for utility, industrial, and commercial/institutional source categories. Additional refinement of the emission factors should include estimates of the particle size distribution of the alkaline particulate emissions to assess the degree to which the particles remain suspended in the atmosphere as a neutralization sink for acid.

VOC EMISSIONS

Although total VOC emissions have been inventoried by the EPA within the National Emissions Data System (NEDS), the Eulerian acid deposition model requires that VOC emission must be disaggregated into refined component species. VOC emissions must be classified by photochemical reactivity with the flexibility of varying the assignments to a class depending upon the modeling chemistry used.

Several major SCCs have either incomplete or no VOC emission factor and speciation data. Available emission factor data are being reviewed to develop the best approch to fulfill the 1985 NAPAP inventory development needs. The program will pursue priorities for VOC emission factor development which reflects NAPAP needs and schedules, the availability of existing data, and the most effective use of project resources to fill current information data gaps.

TABLE 5. ANTHROPOGENIC POINT SOURCE ALKALINE PARTICULATE EMISSION FACTORS

SCC	Generic source description	Units	Percentage of total particulate emission as an alkali metal* Ca	Mg	Na	K	Uncertainty rating**
3-03-023	Iron ore mining and beneficiation	Tons pellets produced	0.49	0.24	0	0	E
3-03-008	Iron and steel industry	Tons produced	3.98	1.66	1.74	5.63	E
3-03-006	Ferroalloy manufacture	Tons produced	1.06	1.45	1.35	0	C
3-04-006	Magnesium production	Tons processed	23.81	13.90	0	0	C
3-05-020	Mining and rock quarrying	Tons raw material	5.77	1.69	0	0	D
3-05-016	Lime manufacture	Tons limestone processed	35.89	5.01	0.50	0	C
3-05-020	Stone processing	Tons raw material	14.38	4.12	0	0	C
3-05-025	Sand and gravel	Tons product	0.65	0.70	0	0	D
3-05-006	Cement manufacture	Tons cement produced	28.50	0.78	0	0	B
3-05-011	Concrete batching	Tons processed	46.00	1.50	0	0	C
3-05-008	Clay products manufacture	Tons input to process	0.85	0.90	0	0	D
3-07-001	Sulfate (Kraft) pulping	Air dry tons unbleached pulp	0.12	0	1.38	0	C
1-01-001 1-01-002 1-01-003 1-02-001 1-02-002 1-02-003 1-03-001 1-03-002 1-03-003 3-90-001 3-90-002 3-90-003	Power generation by utilities	Tons burned	1.62	1.71	0.82	0.44	C
1-01-009 1-02-009 1-03-009 1-05-002-09 3-90-009	Fuelwood combustion	Tons burned	0.05	0.04	0.04	1.95	E
5-02-002-01 5-01-001 5-01-002-01 5-01-005-07 5-02-001	Municipal incineration	Tons burned	0.57	0.14	0.57	0.48	E

*Example: For SCC 3-03-023, emissions of alkaline calcium particulate are calculated as: (particulate per ton pellet produced) x (0.0049). Total alkaline particulate emitted is calculated as: [(particulate per ton pellet produced) x (0.0049)] + [(particulate per ton pellet produced) x (0.0024)].

**Uncertainty Rating of alkali metal content is B = ±25%, C = ±50%, D = ±75%, and E = ±100%.

CONCLUSIONS

Emission factors have been developed for point and area sources of primary sulfate, ammonia, and hydrochloric and hydrofluoric acids for use in the 1980 and 1985 NAPAP emissions inventories. Preliminary alkaline dust emission factors have been developed for anthropogenic point sources from an emissions data set prepared in Canada. Alkaline dust emission factors are needed which consider the particulate size distribution and neutralization capacity of each mass/size fraction. Considerable additions and improvements are needed for VOC emission factors and the level of speciation required to support the development of the Eulerian chemical transformation modules.

NOTE: Although it is EPA policy to utilize metric units, non-metric units have been used in this paper since the NAPAP emission factor file is in non-metric units. For conversion from non-metric units to metric units, note that 1 lb = 0.453 kg that 1 ton (non-metric) = 1 short ton = 2000 lbs = 0.907 metric tons.

REFERENCES

1. Homolya, J. Primary Sulfate Emission Factors for the NAPAP Emissions Inventory. EPA-600/7-85-037 (NTIS PB86-108263), U.S. Environmental Protection Agency, Research Triangle Park, NC, September 1985.

2. Homolya, J. "Stationary Source Emission Factor Development," presented at the First Annual Acid Deposition Emission Inventory Symposium, Raleigh, NC, December 3-4, 1984.

3. Anthropogenic Sources and Emissions of Primary Sulfates in Canada, Ontario Research Foundation, Contract No. 47SS.KE204-1-0318, November 1981.

4. Anthropogenic Sources and Emissions of Alkaline Particulate Matter in Canada, Meteorological and Environmental Planning Ltd., and Ontario Research Foundation, Contract No. 52SS.KE145-3-0403, April 1984.

SIZE SELECTIVE PARTICULATE EMISSION FACTORS
FOR EMISSION INVENTORIES

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FOR EMISSION INVENTORIES

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ABSTRACT

Size specific Particulate Matter emission factors have been developed by the U. S. Environmental Protection Agency (EPA) as a result of source tests performed on eleven major source categories. Within each source category, a number of individual process operations have been characterized by particulate matter size distribution emission rate measurements, along with levels of source activity. In addition, size specific emission factors have been developed for source categories based on information in EPA's Fine Particulate Emission Information System (FPEIS) and in published literature.

INTRODUCTION

In order to implement size specific Particulate Matter National Ambient Air Quality Standards, the U. S. Environmental Protection Agency has been developing data bases for source categories, through the combined efforts of the Office Of Air Quality Planning And Standards (OAQPS) and the Office Of Research And Development (ORD), mainly ORD's Air And Energy Engineering Research Laboratory (AEERL). The primary purpose of this data base development is to produce particle size related emission factors for these source categories to be used principally by States in developing State Implementation Plans when a revised Particulate Matter standard is promulgated by the Agency. These emission factors also can be employed in developing emission inventories.

This paper summarizes briefly the source categories for which particle size emission factors have been developed and made available or are presented in draft reports now undergoing external peer review before their publication in EPA's Compilation Of Air Pollutant Emission Factors, AP-42.

DISCUSSION

Size selective particulate matter emission factor information, resulting from U. S. Environmental Protection Agency's (EPA) emission factor development program, is presented in terms of two efforts; (1) completion of source category reports based primarily on EPA sponsored source testing of selected source categories, and (2) particle size distribution and emission factor information for additional source categories developed from data in the Fine Particulate Emission Information System (FPEIS) and from open literature.

This source categories selected for testing are given in Table 1. Table 2 lists processes within each of these major source categories and presents particle size distribution data from 2.5 to 10.0 micrometers. Fugitive dust particle size emission factor data for urban paved roads, unpaved roads and industrial paved roads are presently available in the Fourth Edition of EPA's Compilation Of Air Pollutant Emission Factors, AP-42. The other source category reports listed in Table 2 are in the process of external peer review before publication of their emission factors in AP-42.

Table 3 presents a list of additional source categories for which particle size data and emission factors are being developed from data in the FPEIS and open literature. These factors are presently in draft form for external peer review.

CONCLUSION

Particle size distribution data and emission factors have been developed for eleven major source categories and a list of sources for which data is available as a result of EPA's efforts. Particle size distribution data range from 2.5 to 10.0 micrometers. Upon publication in AP-42, these data may be used by States for emission inventory and modeling purposes.

TABLE 1. INHALABLE PARTICULATE PROGRAM SELECTED SOURCE CATEGORIES

PAVED ROADS

INDUSTRIAL AND UNPAVED ROADS

IRON AND STEEL

METALLURGICAL COKE

IRON FOUNDRIES

FERROALLOY

PRIMARY AND SECONDARY NONFERROUS METALS

CEMENT AND LIME

ASPHALTIC CONCRETE

KRAFT PULP MILLS

COMBUSTION

TABLE II. PARTICLE SIZE CHARACTERIZATION OF MAJOR SOURCE CATEGORIES PROCESSES

IRON AND STEEL

A source category report for iron and steel presents particle size emission factors for the following:

- °Sinter plant windbox - uncontrolled, and controlled with cyclone, scrubber, electrostatic precipitator (ESP) and baghouse
- °Sinter breakers - controlled with baghouse
- °Blast furnace casthouse - uncontrolled
- °Basic oxygen furnace (BOF) charging and tapping - uncontrolled, and controlled with baghouse
- °Basic oxygen furnace (BOF) refining (O_2 blow) - controlled with scrubber
- °Open hearth - uncontrolled, and controlled with ESP
- °Quelle-Basic Oxygen Process (Q-BOP) - uncontrolled, and controlled refining cycle with scrubber
- °Electric arc furnace - uncontrolled, and controlled with baghouse
- °Hot metal desulfurization - uncontrolled, and controlled with baghouse

METALLURGICAL COKE

The draft report presents particle size emission factors for the following:

- °Coal preheating - uncontrolled, and controlled with scrubber
- °Coal charging sequential
- °Coke pushing - uncontrolled, and controlled with scrubber
- °Push cars in both travel and push modes - controlled with scrubber
- °Coke quenching - uncontrolled (with dirty and clean water), and controlled with baffles
- °Coke oven combustion stacks - uncontrolled

IRON FOUNDRIES

The draft report presents particle size emission factors for the following:

- °Cupola - uncontrolled, and controlled with baghouse and scrubber
- °Metal pouring and cooling - uncontrolled
- °Shakeout process - uncontrolled

FERROALLOY

The draft report presents particle size emission factors for the following:

- °50 percent FeSi open furnace - uncontrolled, and controlled with baghouse
- °80 percent FeMn open furnace - uncontrolled, and controlled with baghouse
- °Si metal open furnace - uncontrolled, and controlled with baghouse
- °SiMn open furnace - uncontrolled, and controlled with scrubber
- °FeCr open furnace - uncontrolled, and controlled with ESP.

PRIMARY AND SECONDARY NONFERROUS METALS

The draft report presents particle size distribution information for the following operations in the primary aluminum, copper, lead, and secondary lead smelting industries.

- °Primary Aluminum
  - Fugitive particulate emissions from a prebake plant - uncontrolled
  - Fugitive particulate emissions from a horizontal Soderberg plant - uncontrolled
  - Prebake reduction cells - uncontrolled
  - Horizontal Soderberg reduction cells - uncontrolled

°Primary Copper

- Multihearth roaster and reverberatory smelter operations - uncontrolled
- Reverberatory smelter operations - uncontrolled and controlled with ESP
- Converter operations - uncontrolled and controlled with ESP
- Reverberatory furnace matte tapping operation fugitives - uncontrolled
- Reverberatory furnace slag tapping operation fugitives - uncontrolled
- Converter slag and copper blow operations fugitives - uncontrolled

°Primary Lead

- Blast furnace - controlled with baghouse
- Blast furnace fugitives - uncontrolled
- Ore storage fugitives - controlled
- Sinter machine fugitives - uncontrolled
- Reverberatory furnace fugitives - uncontrolled
- Dross kettle fugitives - controlled

°Secondary Lead

- Blast furnace - controlled with baghouse
- Blast furnace - (ventilation system fugitives from charging hood, metal and slag tapping hoods) - uncontrolled
- Blast furnace (ventilation system as above) - controlled with baghouse

CEMENT AND LIME

External peer review of the Portland Cement Industry and the Lime Industry Source Category reports have been completed. Reviewer comments are under consideration by EPA and ORD contractors to accommodate these comments and make revisions, where necessary. Particle size distribution and related emissions factors are presented as follows:

°Portland Cement

Wet kiln - uncontrolled, and controlled with ESP

Dry kiln - uncontrolled, and controlled with multiple cyclone and baghouse

Clinker cooler - uncontrolled, and controlled with gravel bed filter

°Lime

Rotary kiln - uncontrolled

Rotary kiln - controlled with cyclone, multiple cyclone, ESP and baghouse

Product loading fugitives - limestone into open trucks and enclosed trucks, and lime into enclosed trucks

ASPHALTIC CONCRETE

External peer review of the source category report is completed. The report is presently being revised to reflect reviewers comments. The report presents particle size emission factors for the following:

°Conventional asphalt plant

Stack emissions - uncontrolled, and controlled with cyclone collector, multiple centifugal scrubber, gravity spray tower and baghouse

°Drum mix plants - uncontrolled, and controlled with baghouse

KRAFT PULP MILLS

External peer review of the source category report is complete. The report presents particle size emission factors for the following:

°Direct contact evaporator (DCE) recovery boiler - uncontrolled, and controlled with ESP

°Nondirect contact evaporator recovery boiler - uncontrolled, and controlled with ESP

°Lime kiln - uncontrolled, and controlled with venturi scrubber or ESP

°Smelt dissolve tank vent - uncontrolled, and controlled with packed tower or venturi scrubber

COMBUSTION

The source category report is currently in external peer review. Particle size emission factors for the following were presented in the draft source category report.

BITUMINOUS COAL

°Dry bottom boiler - uncontrolled, and controlled with multiple cyclone, scrubber, ESP or baghouse

°Wet bottom boiler - uncontrolled, and controlled with multiple cyclone or electrostatic precipator

°Cyclone furnace - uncontrolled, and controlled with scrubber or ESP

°Spreader stoker (overfeed and underfeed) - controlled, and controlled with multiple cyclone

SUBBITUMINOUS (LIGNITE) COAL

°Dry bottom boiler - controlled, and uncontrolled with multiple cyclone, scrubber, ESP or baghouse

°Wet bottom boiler - uncontrolled, and controlled with multiple cyclone or electrostatic precipator

°Cyclone furnace - uncontrolled, and controlled with scrubber or ESP

°Spreader stoker with and without flyash reinjection - uncontrolled, and controlled with multiple cyclone, ESP, or baghouse

°Spreader stoker (overfeed and underfeed) - uncontrolled, and controlled with multiple cyclone

SUBBITUMINOUS (LIGNITE) COAL

°Dry bottom boiler - controlled, and uncontrolled with multiple cyclone

°Spreader stoker - controlled, and uncontrolled with multiple cyclone

ANTHRACITE COAL

°Dry bottom boiler - uncontrolled, and controlled with multiple cyclone

°Stoker - uncontrolled, and controlled with cyclone

RESIDUAL OIL

°Utility boiler - uncontrolled, and controlled with ESP or scrubber

°Industrial boiler - uncontrolled, and controlled with multiple cyclone

°Commercial boiler - uncontrolled

WOOD WASTE

°Bark boilers - uncontrolled, and controlled

°Wood and bark boilers - uncontrolled, and controlled

TABLE III. ADDITIONAL SOURCE CATEGORIES FOR WHICH SOME PARTICLE SIZE DATA AND EMISSION FACTORS ARE AVAILABLE

- Combustion in boilers
  - Bark/Sawdust Boiler
  - Wood Bark/Sawdust Boiler
  - Bagasse Boiler
  - Municipal Garbage Boiler

- Off-Highway Diesel Internal Combustion
- Automobile Spray Booth

- Carbon Black Oil Furnace of Gas Boiler
- Soap and Detergents Spray Dryer

- Sodium Carbonate*
  - Calciner
  - Trona Dryer
  - Dryer
  - Rotary Predryer
  - Bleacher Dryer

- Cotton Ginning*
  - Lint Cleaner Air Exhaust
  - Battery Condenser
  - Saw Gin - Gin Stand and Bale Press
  - Roller Gin - Gin Stand and Bale Press

- Feed and Grain Mills and Elevators
  - Grain Unloading
  - Grain Elevators
  - Agricultural Feed Production

- Ammmonium Sulfate Fertilizer Bed Dryer
- Carob Kibble Dryer
- Cereal Dryer
- Rice Dryer
- Bauxite Processing
  - Ship Unloading
  - Fine Ore Storage Bin

- Secondary Aluminum*
  - Reverberatory Furnace Exhaust
  - Chlorine Demagging

- Steel Foundries
  - Open Hearth Exhaust
  - Shakeout Hood Duct Exhaust

*AP-42 Sections being updated.

TABLE III (CONTINUED). ADDITIONAL SOURCE CATEGORIES FOR WHICH SOME PARTICLE SIZE DATA AND EMISSION FACTORS ARE AVAILABLE

- Lead Battery Manufacture
  - Grid Casting and Small Parts
  - Casting Pot
  - Paste Mixing
  - Lead Oxide Mill
  - Element Assembly Operations
  - Assembly and Ball Mill Processes
  - Reclamation Furnace

- Bricks and Related Clay Products*
  - Screening and Grinding
  - Tunnel Kiln Exhaust
  - Sawdust Firing Brick Kiln

- Coal Cleaning
  - Air Table
  - Thermal Dryer

- Glass Manufacturing*
  - Furnace Exhaust

- Glass Fiber Manufacturing
  - Flame Attenuation Forming Line Exhaust

- Phosphate Rock Processing
  - Calcining
  - Grinding - Ballmill
  - Oil Fired Rotary Dryer
  - Oil Fired Rotary and Fluidized Bed Rock Dryer

- Taconite Ore Processing*
  - Main Waste Gas Stream

- Fluorospar Processing
  - Rotary Drum Dryer

- Petroleum Refining
  - Gas fired Boiler
  - Gasoline Cracking - CO Boiler Exhaust
  - Gas Fired Process Heater

- Wood Products Industry
  - Sanding and Sawmills
  - Belt Sander Hood Exhaust

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*AP-42 Sections being updated.

FIELD MEASUREMENTS STUDIES FOR THE DEVELOPMENT OF POINT SOURCE EMISSION FACTORS

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FIELD MEASUREMENTS STUDIES FOR THE DEVELOPMENT OF POINT SOURCE EMISSION FACTORS

INTRODUCTION

The models under development for prediction of acid deposition require detailed emission data on a number of chemical species. These compounds or elements may act as direct acidic emissions to the atmosphere or may be involved in reactions that produce or eliminate acidic species. Many of the species in question are non-criteria pollutants, and limited information is available concerning emission factors for them. Furthermore, emission data have been requested at levels of temporal and spatial resolution not generally available. A number of projects have been developed to address the need for such data, including literature studies and field tests. This project largely focuses on a series of actual measurements for two particular categories: sulfate emissions from residential and commercial combustion of fuel oil; and emission of chlorine species from coal combustion.

Projects aimed at developing emissions data from literature sources identified gaps in available test data for several categories.[1,2] In the area of sulfate emissions, data on sulfates emitted from the residential and commercial combustion of fuel oil were either absent or based on analytical approaches that have been superseded by more reliable techniques. Data on chlorine emissions from combustion of coal were judged unreliable. It was felt that new measurements should be made employing recent advances in analytical techniques.

TECHNICAL APPROACH

As the emission inventory requirements include spatial and temporal resolution and some measures of addressing uncertainty in the data, a multi-task project was initiated. These tasks include the following:

- Tests for sulfate emissions from two boilers at the Pittsburgh Energy Technology Center (PETC)
- Tests for sulfate emissions at sites external to PETC that are typical of residential and commercial combustion of No. 6 oil
- A literature search and data-base development leading to information on the state or regional distribution of vanadium and nickel in residual fuel oils and the possible impact of these on sulfate emissions
- Tests for chlorine and, incidentally, for fluorine emissions from coal combustion based on the use of a furnace simulator at the Pittsburgh Energy Technology Center (PETC)
- A literature search to develop information on the chlorine content of U.S. coals, and calculation of the equilibrium distribution of chlorine species in flue gas

The sulfate tests at PETC were conducted in two package boilers; a 20-hp fire-tube boiler (700 lb/hr of steam) and a 700-hp watertube boiler

(24,000 lb/hr of steam). These boilers are typical of those package boilers widely used for raising steam in commercial and residential applications. Tests were conducted in the 700-hp facility for high-sulfur No. 6 oils at two excess air levels. Data are reported for four tests in the 20-hp boiler on both No. 2 and No. 6 oil. For the No. 6 oil test, two different excess air levels were studied. All tests were conducted at full load. Samples were drawn using a controlled condensation sampling (CCS) train; analysis of the samples was performed using ion chromatography, with occasional duplicate analyses performed via barium-thorin titration.

Field tests were conducted, through a subcontract to Roy Weston, Inc., at three sites: a residential site with a boiler rated at 145 MBtu/hr; a commercial site with a unit rated at 98 MBtu/hr; and a light industrial facility with a boiler rated at 63 MBtu/hr. Triplicate samples using the CCS method were taken at all locations, and supporting tests using a modified EPA reference method 8 were performed at the commercial site.

Studies have shown that the trace metal content of fuel oils, particularly vanadium, can affect the conversion of sulfur species to SO_3.[3] As extensive data to quantify this dependence are absent, a literature search was made, through a contract with Radian, to develop data on the current and past emissions of vanadium and nickel from combustion sources. These data were used to project sulfate emissions as a function of the variations in vanadium content and fuel use; for one state, these projections were compared, to the sulfate emissions calculated from the best currently available emission factors. It was hoped that this information would contribute to uncertainty estimates in this area.

Chlorine emission tests were conducted in a coal-fired furnace simulator having a firing rate of 500 lb/hr of coal. These tests were piggybacked into a series of tests examining spray dryer/ESP performance in this unit. A sampling train was used that allowed us to collect all chlorine and fluorine species. The train consisted of a glass-lined probe inserted into the stack, a quartz fiber particulate filter, and an impinger train. The solutions in the impinger varied with the test method chosen; both the "LA" method and a method proposed by GCA were used.[4,5] Samples were analyzed using ion chromatography. Additional samples were obtained or samples were split where appropriate and analyzed using the Volhard technique.[4]

Finally, data in the literature indicate that although the chlorine content of United States, coal is low, the actual chlorine content can vary substantially within a given seam. For commercially significant coals in the United States data regarding the chlorine content and its variation were collected from a number of literature sources. In addition, multiphase equilibrium calculations have been performed to provide a basis for comparison with the experimental data.

RESULTS

Table 1 presents the amount of total sulfur converted to sulfates for the tests at PETC. Levels of conversion are typical of equilibrium levels at high temperature with the exceptions of test PS-06. This test was run after 30 hours of operation on No. 6 fuel oil without cleaning the inside of the boiler except for the fire tube itself. Operator comments indicated that the gas passes did have ash deposits in them. With the exception of PS-06, no evidence of conversion due to catalysis by V_2O_5 was found;

however, boiler surfaces were generally clean at the start of each of these tests.

TABLE 1. Test Results from Sulfate Runs

Test	Fuel/%O_2	Sulfate % Total Sulfur	V/Ni (ppm)[a]	Boiler
PS04	0.3%S,No.6F.O./2.2%	1.4	1.4/0.1	20 hp
PS05A	0.35%S,No.6F.O./2.0%	2.1	6.4/8.6	20 hp
PS05B	0.35%S,No.6F.O./3.1%	2.1	6.4/8.6	20 hp
PS06	0.35%S,No.6F.O./3.3%	8.4	6.4/8.6	20 hp
PS07	1.45%S,No.6F.O./2.0%	1.2	94/22.8	700 hp
PS08	1.1%S,No.6F.O./3.0%	1.8	64/12	700 hp

[a] Vanadium and Nickel content of fuel oil (ppm)

A field test program was initiated after reviewing these results. Objectives of the activity were to test typical units, preferably units that had some degree of deposit buildup inside the furnace that might be characteristic of the normal condition of residential and commercial units. In addition, it was planned to conduct tests at one site that had been previously studied and to compare the total measured sulfate from the CCS train to that found by a modified Method 8 train. Table 2 presents data regarding the facilities and operating conditions.

TABLE 2. Site Information for Field Sulfate Runs

Site (---)	Type (---)	Rating (MM Btu/hr)	Capacity Factor (% of Full load)	Excess Air (%)	% S in No. 6 Oil
Starrett City	Residential	145.0	50	40	0.3
Philadelphia State Hospital	Commercial	98.0	21	170	0.5
Wyeth Laboratories	Light Industrial	63.0	75	25	1.0

The field tests were conducted in the spring and summer, accounting for the low load factors at the residential and commercial sites and accounting for the high excess air at the commercial site. The residential boiler had been cleaned immediately before to the start of testing; the boiler at the commercial site had fired fuel oil for 72 hours before the testing; the industrial boiler had not been cleaned in the recent past. Table 3 provides a summary of the results.

TABLE 3. Test Results from Field Tests

Site	Method	V/Ni[a]	% S from Fuel as Primary Sulfate
Starrett City	CCS[b]	14.9/16.4	1.5% (37% as acid)
Philadelphia State Hospital	CCS	15.5/30.8	0.95% (46% as acid)
Philadelphia State Hospital	IPA[c]	15.5/30.8	1.5%
Wyeth Laboratories	CCS	16.1/27.5	3.8% (82% as acid)

a Vanadium and Nickel (ppm)
b Controlled Condensation Sampling Train
c Modified EPA Reference Method 8

These data that the conversion to sulfate was 2.0% or less for the lower load tests with boilers that had been cleaned recently. Only for the Wyeth Laboratories test was a higher sulfate conversion noted, and all of this increase appeared as acid. In addition, the results from the modified Method 8 test were consistently higher than from the tests using the CCS train.[6]

Some results from the vanadium data base are shown in Figure 1. Calculated emissions are shown by state for the years 1950 and 1980. Only Ohio, Oregon, Pennsylvania, and Rhode Island have shown a decrease. Other states that now have reasonably large emissions have shown a significant increase over the 1950 numbers. Among these are Alabama (4-fold), Delaware (7-fold), Florida (6-fold), Louisiana (18-fold), Mississippi (88-fold), and North Carolina (9-fold). Each of these six states emits over 100,000 kilograms of vanadium per year (1980) and each has shown more than a 400% increase in such emissions since 1950.[7]

Results of the chlorine and fluorine tests have been reduced to the form of emission factors, and are presented in Table 4 as pounds per million Btu per percent of the element in the fuel. The values calculated from the emission tests in the stack are in reasonably good agreement with the quantities of each element that entered in the fuel. Furthermore, the results indicate that essentially all the chlorine and fluorine are present as HCl and HF.

Equilibrium calculations were performed using a multiphase equilibrium program available at PETC. These calculations predict that HCl would be the dominant gas phase species (by over two orders of magnitude) at the temperatures of interest. These calculations agree with earlier studies.[8,9,10]

TABLE 4. Calculated Emission Factors

Element (X)	Calculated Emission Factors		
	lb/($\bar{M}$Btu)[a]	lb/($\bar{M}$Btu x %X)[a]	lb/($\bar{M}$Btu x %X)[b]
Cl	4.4×10^{-2}	0.69	0.76
F	4.4×10^{-3}	0.87	0.71

[a] Calculated From Emission Tests
[b] Calculated From Fuel Compositions and Feed Rate

CONCLUSIONS

In general, the results of all the sulfate tests indicate low conversions of sulfur to primary sulfate. However for two tests, where the boiler may have contained significant deposits, higher conversions were noted. Data have been developed for a range of unit sizes, operating conditions, and fuel properties.

Chlorine and fluorine emissions follow the concentration of these elements in the fuel; HCl and HF are the dominant gas-phase species.

References

1. D. Misenheimer, R. Sommer, M.R. Clowers, and A.S. Werner, "Assessment of Ammonia, Hydrogen Chloride, Hydrogen Fluoride Emission Factors for the NAPAP Emission Inventory," GCA Corporation EPA Contract No. 68-02-3168, TASK No. 90, Revised Draft Final Report, March 1985.

2. J. Homolya, "Primary Sulfate Emission Factors for the NAPAP Emissions Inventory," Radian Corporation, EPA Contract No. 68-02-3994, Task No. 5, Revised Draft Final Report, July 1985.

3. Workshop Proceedings on Primary Sulfate Emissions from Combustion Source, Volume 2, U.S. Environmental Protection Agency, NTIS PB-287 436, August 1978.

4. J. Driscoll, Flue Gas Monitoring Techniques, Ann Arbor Science Publishers, Inc., Ann Arbor, Mich.

5. D.A. Stern, B.M. Myatt, J.F. Lachowski, and K.T. McGregor, "Speciation of Halogen and Hydrogen Halide Compounds in Gaseous Emissions," presented at the Ninth Annual Research Symposium, Land Disposal, Incineration, and Treatment of Hazardous Waste, Ft. Mitchell, Ky., (May, 1983).

6. "Characterization of Primary Sulfate Emission from a Residential and a Commercial Boiler Firing No. 6 Fuel Oil," draft final report by Ray F. Weston under Subtask 7.9, U.S. DOE Contract DE-AC-22-84PC75571 (August 1985).

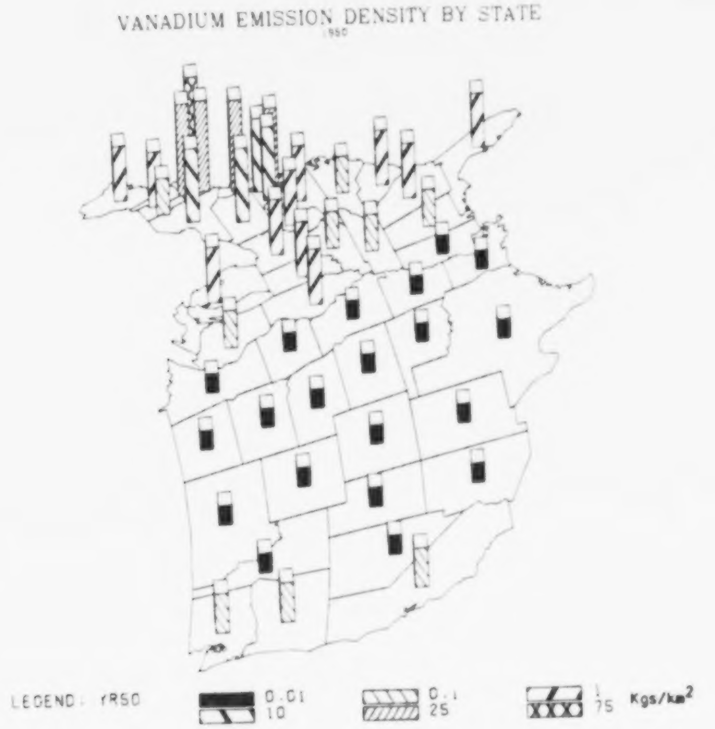
7. S. Kulkarni, C. Arnold, J. Homolya, and J. Dickermann, "Vanadium Nickel and Primary Sulfate Emissions From the Combustion of Fossil Fuels in the United States of America," draft final report by Radian Corporation under U.S.DOE Contract No. DE-AC-84PC71505 (August 1985).

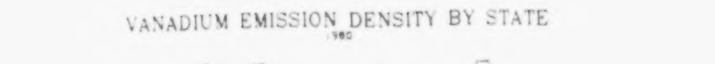
8. T.L. Iapalucci, R.J. Demski, and D. Bienstock, "Chlorine in Coal Combustion," U.S. Department of the Interior RI7260 (May 1969).

9. M.J. Hodges, W.R. Ladner, and T.G. Martin, "Chlorine in Coal: A Review of Its Origin and Mode of Occurrence," Journal of the Institute of Energy, pp. 158-169 (September 1983).

10. W.D. Halstead and E. Raask, "The Behavior of Sulphur and Chlorine Compounds in Pulverized-Coal-Fired Boilers," Journal of the Institute of Fuel, pp. 344-349 (September 1969).

Figure 1. Computed Vanadium Distribution
For 1950 and 1980


VANADIUM EMISSION DENSITY BY STATE
1950
LEGEND: YR50
0.01
10
0.1
25
1
75
Kgs/km²


VANADIUM EMISSION DENSITY BY STATE
1980
LEGEND: YR80
0.01
10
0.1
25
1
75
Kgs/km²

VOC COMPOSITION OF AUTOMOTIVE EXHAUST AND SOLVENT USE IN EUROPE

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VOC COMPOSITION OF AUTOMOTIVE EXHAUST AND SOLVENT USE IN EUROPE

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ABSTRACT

Compositions of automotive exhaust hydrocarbons and aldehydes are presented at a level of detail relevant for input into photochemical air pollution and acid deposition models. Data were compiled from literature for gasoline-, diesel- and LPG-powered vehicles without exhaust controls. For gasoline-derived compositions, data were converted to separate profiles for saturates, unsaturates and aromates. Relations between gasoline composition and exhaust composition were used to combine the average profile compositions to one average overall composition. A comparison with fuels measurements was made.

An attempt has also been made to estimate an average solvent consumption resp. emission pattern for Western European countries. Since data appeared to be very scarce, all available material both inside Europe and abroad was used for a first approximation. An average mass rate per capita and an average composition is proposed. Spatial distribution criteria are suggested. The results should be used as default values anticipating more reliable data.

VOC COMPOSITION OF AUTOMOTIVE EXHAUST AND SOLVENT USE IN EUROPE

INTRODUCTION

The two largest contributors to total anthropogenic VOC emissions in most European countries are non-catalyst automobiles and the evaporation of solvents (ca. 50 percent and ca. 35 percent of total VOC emissions respectively). For the purpose of modeling photochemical air pollution and acid deposition, it is necessary to know the composition of these pollutants as detailed as relevant. Modeling large areas in Europe implies the possibility that nationally different emission factors have to be used. While differences in vehicle fleets as well as driving habits can reasonably be taken into account in estimating mass emissions, their influences on composition are not known. When solvent emissions have to be assessed, considerably larger problems must be faced as both mass emission rates and compositions are sufficiently known for only a few countries.

The need of widely applicable emission factors being apparent, an attempt has been made and is presented in this paper to quantify:

a. exhaust hydrocarbon composition from gasoline-powered vehicles with only fuel composition as a variable,

b. exhaust hydrocarbon compositions of diesel- and LPG-powered vehicles,

c. an average per capita mass emission factor for solvent use,

d. an average composition of solvent emissions

and, finally, to draw up spatial distribution criteria for solvent emissions.

DISCUSSION

1. Exhaust Emissions

For gasoline-powered vehicles, the literature was searched for test data satisfying the following demands: a) test vehicle(s) without exhaust control, b) VOC-analysis comprising at least main components, c) complete (or nearly complete) mass balance. Eleven investigations answered these demands (Table 1).

All data were converted to percent by weight to facilitate combination with mass emission factors. As could be expected, averaging absolute fractions was not of much use. Therefore, composition profiles were calculated for saturates, unsaturates and aromates. These resulted in applicable averages (Tables 2a and 2b).

Next the profiles had to be integrated to one average composition. Only for aromates a useful correlation between fuel- and exhaust composition appeared to exist (Figure 1). It has been shown, however (12, 13), that there is a relation between fuel aromates and exhaust olefines (Figure 2). With this, exhaust composition can be estimated by the aromatic content of the fuel (Figure 3). Finally, a correction for oxygenates is needed. Oxygenate content and composition, as well as diesel- and LPG-exhaust composition were estimated from literature data satisfying the same above-mentioned demands (Tables 3-5). In contrast with gasoline-exhaust composition, useful material was scarce; for diesel fuel only six more or less incomplete data sets were found, for LPG only two (20, 22).

Since only one report about field measurements was found that could be used as a comparison (21), an average road traffic exhaust composition was calculated for that purpose (Netherlands, 1978; Table 6). In this table, a comparison based on a reactivity scheme has been added because this is what ultimately is of interest.

2. Solvent Emissions

Assuming that a society's solvent consumption, with respect both to quantity and substances used, depends on the level of industrialization and material development, it can be expected that comparable emission patterns exist in several European countries. This was checked by collecting as much data as possible about solvent emissions from different countries and taking averages wherever it made sense (Tables 7 and 8). Due to divergencies among data and uncertainties about representativeness the results should be characterized as default values only. Another approach, in progress now, is the assessment of mass balances to which solvent consumption factors are applied. It is not certain yet whether this will give more reliable results.

The geographic distribution of solvent emissions is complex because a comprehensive source type-substance matrix is involved. In view, however, of the very large number of small sources and the relatively few large ones, it seems justifiable to distribute all emissions proportional to population density and make corrections for large area sources afterwards. This approach is valid only for meso- and macro-scale modeling.

CONCLUSIONS

An average composition of road traffic exhaust (dominated by gasoline powered vehicles), based on a literature survey comprising old as well as recent investigations, has a variation that is considered appropriate for air pollution modeling. Taking into account the scarce data about the influence of vehicle speed, vehicle age and ambient temperature on composition, indicating that these influences cause no larger variations than those presented in this study, it seems reasonable to assume that no better input data can be expected to be available for modeling than these.

The quality of emission factors for solvent use can not easily be estimated since there is insufficient knowledge about the reliability of individual national data. Analysis of national mass balances may possibly improve the factors.

LITERATURE

1. J.D. Caplan, SAE Progr. Tech. Ser. 12 (1967) 20-31.
2. M.W. Jackson, ibid, 241-67.
3. I.D. Lytollis, MIRA Report Nr. 1971/1 (1970).
4. W.E. Morris, K.T. Dishart, ASTM STP 487 (1971) 63-101.
5. K. Schofield, Env. Sci. and Tech. 8, 9 (1974) 826-34.
6. L.J. Papa, SAE Progr. Tech. Ser. 14 (1971) 43-65.
7. B. Dimitriaded *et al.*, Bur. of Mines Rep. of Inv. 7700 (1972).
8. S. Yanagihara, Proc. 16th FISITA-Congress, Tokyo 1976, 2-149-56.
9. F.M. Black, L.E. High, EPA-600/2-80-085 (PB 203136), JAPCA 30, 11 (1980) 1216-21.
10. P.F. Nelson, S.M. Quigly, Atm. Env. 18, 1 (1984) 79-87.
11. R. Steenlage, R.C. Rijkeboer, MT-TNO Rep. Nr. G 1115 (1985).
12. E.E. Wigg, *et al.*, SAE-Paper 720251 (1972).
13. K. Hosaka, *et al.*, SAE Tech. Paper Ser. 780625 (1978).
14. P.M. Carey, EPA-AA-CTAB-PA-81-11 (PB 82-118159) (1981). (a) 4 LDV's (1970); FTP; DNPH. (b) 10 LDV's (1970-1973); FTP; DNPH. (c) AMC Pacer (1977); FTP; DNPH (average value of five types of malfunction).
15. See 12. 1970 repr. test vehicle; FTP; MBTH (total), DNPH (ind.)
16. P.E. Oberdorfer, SAE Progr. Tech. Ser. 14, 32-40 (1971).
17. C.S. Wodkowski, *et al.*, West Coast APCA Mtg., San Francisco 1970 (cit. from Vapor-Phase Organic Pollutants, EPA 600/1-75-005, PB 249357); a = 30 mph, b = FTP).
18. M.F. Fracchia, *et al.*, Env. Sci. and Tech. 1, 11 (1967) 915-22.
19. Nat'l Res. Council, EPA 600/6-82-002 (PB 82-180498); a = idle, b = FTP.
20. R. Steenlage, R.C. Rijkeboer, MT-TNO Rep. Nr. G 1115 (1985) (Dutch. Org. for Appl. Sci. Res.); Opel Ascona 1.6 S (1983), EEC-cycle, MBTH; a = optimal, b = rich.

21. R. Guicherit, Th.R. Thijsse, IMG-TNO Rep. Nr. G 1198 (1982).

22. K.E. Egeback, _et al._, Nat'l Swedish Environ. Protect Board Rep. Nr. 1635 (1983).

23. Umweltbundesamt Berlin.

24. CITEPA, Paris.

25. K.A. Brice, R.G. Derwent, Atm. Env. _12_ (1978) 2045-54.

26. Project Emission Registration, Min. of Housing, Physical Planning and Environment (Netherlands).

27. Swedish Environ. Res. Inst., Stockholm.

28. EPA, Office of Air Quality Planning and Standards.

29. Environ. Protect. Progr. Dir., Report EPA 3-EP-83-10 (1983).

30. Photochemical Smog, OECD, Paris 1982.

31. R. Schaaf, Wasser, Luft and Betrieb, 1984-3, 36-8.

32. C. Morley, Shell Res. Ltd., Thornton Res. Centre, England; paper for submission to Atmospheric Environment (1981).

33. P.F. Nelson, _et al._, Atm. Env. _17_, 3 (1983) 439-49.

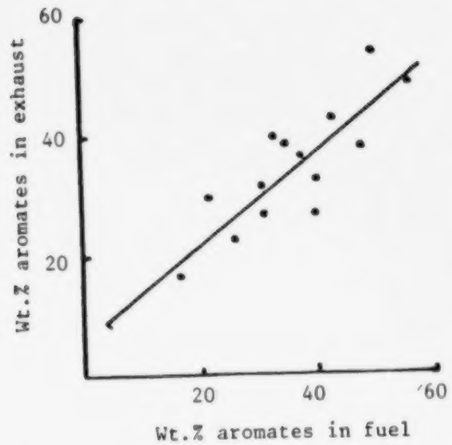

Fig.1

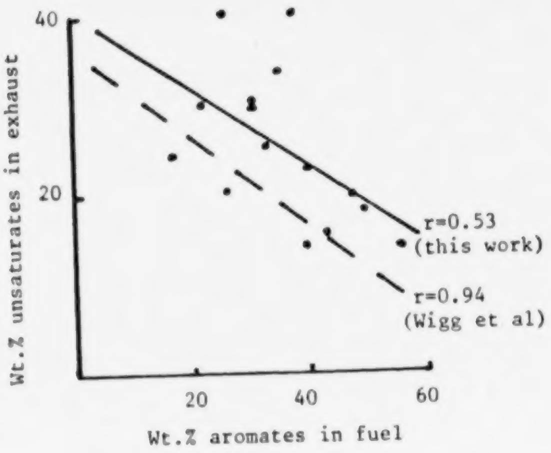

Fig.2

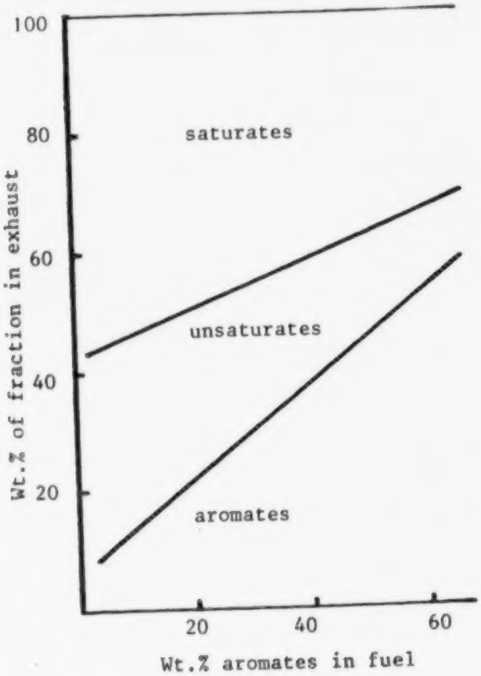

Fig.3

Table 1

Investigations of automotive exhaust; gasoline powered vehicles; experimental data.

Lit. nr.	Test vehicle(s)	Test procedure	Gasoline composition (wt.%)			Approx. year of investig.
			sat.	unsat.	arom.	
1	no data					1967
2	0.63 l single cyl. motor; compression ratio = 8.9/1		55	10	35	1967
3	1 of 4 cyl. of 1.3 l motor; compression ratio = 8.5/1		46	16	38	1970
4	Automobile without control system		59	10	31	1968
5	No data; "representative composition"					1971
6	"62 privately owned and operated automobiles". Composition of one of these is given.	Calif. cycle				1970
7	1969 Valiant; 3.8 l	7-mode Calif. cycle. Each fuel: rich, resp. lean; with, resp. without vacuum spark advance	a) 63.5 b) 57 c) 48	14.5 10 2	22 33 50	1971
8	"pre-emission controlled car with retrofit device of retarding ignition timing"	Hot 10-mode, 6 cycles	a) 51 b) 71 c) 62 d) 59.5 e) 41 f) 40 g) 33	6 12 12 0.6 19 12 11	43 17 26 40 40 48 56	1975
9	1963 Chevrolet 283 V8	FTP	69	5	26	1979
10	67 automobiles representative for Sydney fleet. Ages: < '72 - > '78. Weighed average composition given.	1975 FTP	63.5	6	30.5	1980
11	Opel Ascona 1.6 S (1983)	EEC-cycle, rich and optimal; 90 km/h opt. 120 km/h opt.				1984

Table 2a Investigations of automotive exhaust; gasoline powered vehicles. Hydrocarbon analyses (wt.%)

Hydrocarbon	1	2	3	4	5	6	7			8							9[3]	10[4]	11
							a	b	c	a	b	c	d	e	f	g			
saturated																			
methane	22.0	16.0	20.1	12.8	23.7	19.1	14.8	12.9	15.7	14.6	22.9	22.4	28.0	40.3	28.6	31.1	28.5	14.8	21.7
ethane	2.8	4.0	4.7	2.7	3.6	1.7	1.6	1.5	1.5								5.4	2.8	4.3
propane	-									0.2	0.3	0.4	0.5	0.3	0.4	0.5		0.2	0.3
butanes	4.7	5.1	12.0	10.3		11.0	5.7	6.0	11.6	3.7	4.6	3.9	2.6	4.7	4.1	2.7	10.7	7.7	6.1
pentanes	13.3	14.9	19.8	16.0		14.0	12.7	16.9	13.7	22.7	10.7	13.6	15.1	18.2	18.4	19.6	9.0	21.1	25.2
hexanes	9.4	12.4	17.4	19.1		16.3	24.8	19.5	11.9	35.4	15.0	18.5	22.8	19.8	27.6	26.2	14.8	22.0	2.0
heptanes	7.5	9.4	13.5	13.4		12.3	14.7	10.4	11.6	8.7	10.3	6.2	7.4	6.8	9.5	10.0	9.7	11.7	9.7
octanes	40.3	35.9	10.5	}25.7		20.8	10.7	9.3	21.4	10.2	34.1	30.3	20.8	5.1	6.4	6.3	16.7	10.1	27.2
C ≥ 9 saturates		2.2	2.0			4.8	15.0	23.6	12.6	4.5	2.1	4.7	2.8	4.8	5.0	3.6	5.2	9.6	3.5
wt. % abs.	30.8	27.8	23.2	42.8	34.5	44.4	40.3	35.0	28.0	41.7	58.8	56.8	53.4	50.6	42.1	37.4	37.3	38.8	27.0
unsaturated																			
ethylene	21.5	18.3	33.6	29.9	22.4	25.5	29.3	24.6	22.8	33.2	30.7	32.9	39.1	31.9	36.2	39.0	34.4	31.2	28.2
acetylene	15.9	13.1	24.5	19.6	25.8	35.7	22.4	21.4	27.8	24.9	25.4	28.1	24.3	23.8	39.7	29.4	24.6	27.5	42.4
propylene	12.6	15.8	13.1	18.5	17.5	11.8	11.3	10.4	15.4	13.1	23.5	15.8	13.8	16.7	8.9	11.6	14.8	13.6	13.9
propadiene		2.6	}3.6		2.3	0.5	0.7	0.5	1.0								0.8		1.3
methylacetylene	1.8					1.0	1.2	1.3	2.1								1.9	1.2	
1-butenes	}12.5	}16.0	}8.5	10.6	}12.9	5.7	4.2	3.5	6.4	3.7	4.0	3.7	3.7	4.1	3.1	2.6	6.4	6.2	12.2
1,3-butadiene							2.8	2.5	1.6								4.3		1.3
2-butenes	2.5	3.8	3.7			2.9	2.0	2.8	3.4	2.4	2.5	2.8	1.9	6.2	1.3	1.6	2.1	3.1	0.7
1-pentenes	3.1	3.5	3.7			3.6	2.8	4.2	2.5	5.1	3.4	3.7	1.6	3.3	1.4	2.3	6.0	1.5	
2-pentenes	7.0	6.9	4.6	}21.4		7.9	4.2	5.9	5.2	10.3	4.0	4.4	3.8	7.6	2.1	3.0	2.0	3.4	
1-hexenes	4.7	4.2	1.4			1.2	3.6	2.9	4.2	3.5	1.3	2.0	4.0	1.6	2.1	2.6	1.4		
2- and 3-hexenes	3.8	3.6	1.8			2.7	3.3	4.3	1.5	3.8	1.6	2.5	1.3	2.5	1.6	2.9	1.1	}12.3	
C ≥ 7 unsaturates	14.6	12.2	1.5			1.5	12.2	15.7	6.1		3.6	4.1	6.5	2.3	3.6	5.0	0.2		
wt. % abs.	32.0	33.7	40.6	30.3	37.1	24.9	30.2	25.3	18.0	15.5	24.6	20.4	14.1	22.8	19.9	13.9	40.3	29.5	25.3
aromatic																			
benzene	8.2	7.2	11.3	11.0	12.5	8.3	5.7	2.6	2.9	14.9	14.7	15.0	12.3	16.2	14.8	46.2	17.6	14.0	10.3
toluene	47.2	46.7	20.1	32.5	40.8	20.8	14.5	10.0	34.7	32.0	26.3	27.3	32.3	31.1	36.2	21.5	22.7	29.2	23.9
xylenes	11.2	11.0	23.2	22.5	14.0[2]	21.4	18.5	19.6	25.7	23.7	23.5	24.0	22.2	24.4	24.3	15.8	26.6	25.5	34.5
ethylbenzene	3.0	3.5	5.4	7.5	4.0	4.3	3.6	3.5	5.8	5.4	6.3	5.8	6.1	6.5	7.1	4.1	5.2	5.4	10.1
styrene							1.3	0.9	0.9										2.8
1,2,4-trimethylbenzene		7.8[1]	9.8	8.5		8.3	9.8	9.6	7.5	5.4	5.6	4.5	4.4	2.4	3.6	2.7	10.7	6.0	}8.1
1,3,5-trimethylbenzene	1.2	1.5	2.9			2.5	2.3	1.9	1.5	2.2	2.5		1.3	1.8	1.0	0.7	4.2	2.3	
1,2,3-trimethylbenzene			1.3	}18.0		0.7	}11.4	}11.9	}10.6	3.9	1.5	2.0		1.0	1.1	1.5	}10.4	}9.7	
other C_9-aromates	}29.2	8.6	12.0			11.5				6.4	8.6	6.8	7.0	5.3	6.7	3.8			7.2
C ≥ 10 aromates		13.7	14.0			22.2	32.9	40.0	10.4	6.1	11.0	14.6	14.4	11.3	5.2	3.7	2.6	7.9	0.3[5]
wt. % abs.	37.2	38.5	36.2	26.9	28.4	30.7	29.5	39.7	54.0	42.8	16.6	22.8	32.5	26.6	38.0	48.7	22.4	31.7	47.7

[1] + C_{10} arom.
[2] + styrene
[3] some peaks interpreted with results of 7
[4] 6% methane and 4% unsat. C_6 + assumed
[5] naphtalene

Table 2b

Investigations of automotive exhaust; gasoline powered vehicles

Average values (wt. %)

Hydrocarbon	average	%σ	average (excl. 2σ)	%σ	n	adopted average
saturated						
methane	21.6 ± 7.3	34	20.5 ± 5.9	29	18	19
ethane	3.1 ± 1.3	44	3.1 ± 1.3	44	12	3
propane	0.3 ± 0.1	33	0.3 ± 0.1	33	9	0.5
butanes	6.5 ± 3.2	49	6.5 ± 3.2	49	18	
pentanes	16.4 ± 4.2	26	15.9 ± 3.7	23	17	
hexanes	18.6 ± 7.5	41	18.6 ± 5.2	28	16	
heptanes	10.2 ± 2.4	23	10.2 ± 2.4	23	18	
octanes	18.6 ± 11.4	61	18.6 ± 11.4	61	18	
C9 + saturates	6.2 ± 5.9	95	5.5 ± 3.9	70	15	
	101.5		99.2			
C_4 + saturates	76.7 ± 7.3	9	77.6 ± 6.1	8	17	77.5
						100
unsaturated						
ethylene	29.7 ± 5.9	20	29.7 ± 5.9	20	19	29.5
acetylene	26.1 ± 7.2	28	25.2 ± 6.2	25	18	25
propylene	14.3 ± 3.3	23	13.8 ± 2.5	18	18	14
propadiene	1.2 ± 0.8	67	1.2 ± 0.8	67	8	1
methylacetylene	1.6 ± 0.5	31	1.6 ± 0.5	31	7	1.5
1-butenes	5.3 ± 2.8	52	4.9 ± 2.1	43	14	6
1,3-butadiene	2.5 ± 1.2	47	2.5 ± 1.2	47	5	2.5
1-pentenes	3.2 ± 1.2	39	3.0 ± 1.0	34	15	3
2-pentenes	5.1 ± 2.3	45	4.8 ± 1.9	40	15	4
1-hexenes	2.7 ± 1.2	46	2.7 ± 1.2	46	15	2.5
2- and 3-hexenes	2.6 ± 1.0	41	2.6 ± 1.0	41	15	2.5
C_{7+} unsaturates	6.4 ± 5.2	81	6.4 ± 5.2	81	14	6
	100.9		100.9			100
aromatic						
benzene	12.9 ± 9.1	71	11.1 ± 4.5	40	18	12
toluene	28.9 ± 9.9	34	28.9 ± 9.9	34	19	29
xylenes	23.1 ± 6.8	29	22.5 ± 6.1	27	19	23
ethylbenzene	5.8 ± 2.1	37	5.3 ± 1.6	30	19	5
styrene			1		4	1
1,2,4-trime.-benzene	6.6 ± 2.8	42	6.6 ± 2.8	42	15	6.5
1,3,5-trime.-benzene	2.0 ± 0.9	44	1.8 ± 0.7	36	14	2
1,2,3-trime.-benzene	1.6 ± 1.0	61	1.3 ± 0.4	32	7	1.5
other C9 aromates	7.6 ± 2.3	30	7.6 ± 2.3	30	11	8
C_{10+} aromates	14.0 ± 10.5	75	12.1 ± 7.9	65	15	12
	102.6		98.3			100

Table 3

Oxygenates in automotive exhaust; gasoline-powered vehicles (wt. %)

Component	14			15	16	17		18	19		20		average (excl. 2σ)	%σ	adopted average
	a	b	c			a	b		a	b	a	b			
formaldehyde		86	74.2	48.7	48.5	36.9	42.0	53.0	53.9	49.3	50.0	46.0	50.3 ± 9.8	20	50
acetaldehyde		13.5	19.6	9.3	7.3	12.9	6.7	9.1	9.2	13.4			11.2 ± 4.0	36	10
acetone			5.2	}22.2	0.5	8.2	0.8	3.0	9.0	23.0			4.5(0.5-9)		5
propionaldehyde															
acroleïn				} 5.0	12.7		3.0	8.7					8.1(3-12.7)		
butylaldehydes					0.7	4.4	1.5	3.0	3.0	2.0			2.4 ± 1.3	54	
hexanaldehyde			1.2												
benzaldehyde				12.4	20.8	7.2	11.6	11.1	11.2	10.2			12.1 ± 4.2	35	10
tolualdehyde				12.3		14.5	7.5						11.4(7.5-14.5)		10
other aldehydes				0.9	9.5	15.9	26.9	12.1	13.7	2.1					15
total oxygenates	2.0	5.8	1.9								2.3	1.6	2.7 ± 1.7	64	2

See list of references for experimental data

Table 4

Composition of diesel exhaust

Component	average	adopted average
methane	3.7 ± 1.5	4
ethane	0.5	0.5
saturates C_{13}^{-}	50	50
ethylene	11.0 ± 3.4	11
acetylene	3.2 ± 1.6	3
propylene	3.4 ± 1.0	3.5
1-butenes	1.5 ± 0.5	1.5
pentenes	0.7	0.5
unsaturates C_{13}^{-}	2	2
benzene	2.6 ± 1.1	2.5
aromates C_{12}^{-}	8	8
formaldehyde	5.9 ± 2.5	6
aliph. aldehydes C_4^{-}	4	4
unsat. aldehydes C_3^{-}	0.5	0.5
acetone	2 (0.6-3.6)	2
organic acids C_2^{-}	1	1

Table 5

Composition of LPG-exhaust

Component	average	adopted average
methane	8.9 (1.3-16.4)	9
ethane	2.7 ± 0.8	3
propane	34.5 ± 6.0	35
ethylene	15.6 ± 4.9	15
acetylene	21,8 ± 5.7	22
propylene	7.6 ± 2.1	8
benzene	0.15(0.03-0.4)	
toluene	0.3 (0.06-0.5)	
xylenes	1.5 (0.06-3.5)	1.5
formaldehyde	3.4 ± 1.5	4
acetaldehyde	0.6	1
organic acids	1.7 ± 1.0	1.5

Table 6

Road traffic exhaust composition (wt. % and mole/kg[2])

	calculated [1]	measured (21)
methane	8.0	7.2
ethane	1.3	0.6
propane	1.2	0.6
C_4-C_{10} saturates	29.8	32.3
C_{11+} saturates	5.5	2.2
ethylene	7.9	6.6
acetylene	6.3	4.8
propylene	3.6	2.6
propadiene	0.2	
methylacetylene	0.3	
1-butenes	1.4	1.0
1,3-butadiene	0.5	
2-butenes	0.5	0.4
1-pentenes	0.7	1.2
2-pentenes	0.9	1.3
1-hexenes	0.5	2.4
2- and 3-hexenes	0.5	
C_{7+} unsaturates	1.5	0.7
benzene	3.2	3.9
toluene	7.2	9.9
xylenes	5.7	6.4
ethylbenzene	1.2	2.3
styrene	0.3	0.8
1,2,4-trime.-benzene	1.6	2.3
1,2,5-trime.-benzene	0.5	1.3
1,2,3-trime.-benzene	0.4	0.6
other C_9-aromates	2.0	2.8
C_{10+} aromates	3.9	3.7
formaldehyde	1.6	1.1
other oxygenates	1.8	1.0
OLE	1.4	1.1
PAR	32.7	31.1
TOL	0.9	1.3
XYL	1.2	1.4
FORM	1.5	0.7
ALD	0.6	1.0
ETH	2.8	2.4
UNR	11.1	10.0

[1] 30% aromates in gasoline, gasoline exhaust: 86%, diesel exhaust: 11%, LPG exhaust 3% (Dutch 1978 average).

[2] Chemical bond species (CBM-X, SAI)

Table 7

VOC-emissions from solvent use
(kg/cap. year)

	23 FRG 1978-'80	24 France 1979	25 UK 1975	26 Netherlands 1981	27 Sweden 1984	28 USA 1982	29 Canada 1978	default value
INDUSTRIAL								
paint application	3.25			(2)[2]	2.4	6.8		2
degreasing	2.3[1]	1.65	0.45		1.2	1.6	2.25	1.6
printing	1.15	0.65		0.55	0.85	1.3		1.0
chemical industry				1.3	0.35	4.1		
storage & handling				0.15				
metal products manuf.				0.7				
glues, adhesives	0.8							
other	0.3			2.4				
total from industry	7.8			5.1	4.8	13.8		
NON-INDUSTRIAL								
paint application	2.6		2.6	2.7	1.2	2.85		2.5
chemical cleaning	0.25	0.55	0.66	0.25	0.18	0.95	0.7	0.5
consumer products				3.5	3.8[3]	3.5	0.9	3.5
- a.w. aerosol cans				1.25				
total non-industrial				6.45	5.2	7.3		6.5
total emissions	13.7	13	5.5	11.6	10	21.1	5.6	12
total paint application	5.85	3.7		4.7	3.6	9.65		4.5
OECD-estimation 1975[4]	13.2	12.2	5.5[5]	11.4	12.1	29.5	10.0	
1985	18.8	17.5	7.4	15.3	16.1	40.4	13.5	

[1] or 1.9

[2] included in industrial emissions

[3] OECD-figure

[4] Ref. 30

[5] from ref. 25

Table 8

Compositions of VOC-emissions from solvent use
(wt. %)

	31 FRG 1982	24 France 1978	32 OECD Europe2)	26 Netherlands 1981	28 USA 1982	33 Sydney	default value
sat. hydrocarbons C_{4+}	19.2	27	36.3	42.5	59.3	25.7	32
toluene	5.4		5.1	>6.6		18.4	6
xylenes	3.9		4.0	>3.1		10.6	4
other aromates	1.6			9.8	1.6	16.6	5
total aromates	10.9	24	9.1	19.5		45.6	
methylalcohol	3.8		1.2	1.3	5.0		2
ethylalcohol	3.3		6.1	8.6	5.3	13.7	6
isopropylalcohol	1.4	(6)	6.7		2.6		5
butanoles	6.7			3.8	1.5	1.3	
cellosolves	5.3		6.4		4.7		
other alcohols		24[1]			0.2		5
total alcohols	20.5		20.4	13.7	19.3	15.0	
acetone		(6)	6.1	2.3	3.2	1.6	4
methylethylketone			3.2		4.1	2.9	3
methylisobutylketone			2.1	2.0	1.0	1.0	2
other ketones					0.3		
total ketones	5.4		11.4	4.3	8.6	5.5	
acetates	7.1	9	5.1		2.8	2.6	6.5
other esteres	0.5						
total esters	7.6		5.1	3.9	2.8	2.6	
ethers	11.9			1.9			2
methylene chloride	6.0		5.5	4.6		1.8	5
trichloroethylene	2.2		4.4	1.3	1.3	1.9	2.5
perchloroethylene	4.4		4.8	3.5	4.0	0.4	4
other Cl-hydrocarbons	4.9		3.0	1.0	1.6	1.5	2.5
total Cl-hydrocarbons	1.75	16	17.7	10.4	6.9	5,6	
Cl-F-hydrocarbons	3.8			3.6			3.5
other solvents	3.2			0.2	1.5		

[1] incl. ethers

[2] Non-reactive compounds not included. Added for this study: 300 kt acetone, 60 kt methylalcohol, 270 kt methylene chloride, 150 kt trichloroethane.

SESSION 4: DEVELOPMENT OF EMISSION INVENTORIES FOR NATURAL SOURCES

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MEASUREMENT OF BIOGENIC SULFUR FLUXES AT THREE SITES IN THE EASTERN UNITED STATES

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MEASUREMENT OF BIOGENIC SULFUR FLUXES AT THREE SITES IN THE EASTERN UNITED STATES

INTRODUCTION

An assessment of the significance of natural biogenic sulfur sources compared to anthropogenic sources as potential contributors to the acidification of rainfall requires measurement of fluxes at or below 10 ng S/m^2 min over large geographic areas. To date, such an assessment has rested almost entirely on one study completed in 1979, by Adams, et al. (1). In view of the importance attached to these measurements, the analytical difficulties recognized by previous workers, and recent improvements and advances in measurement technology for trace sulfur species, a reassessment of the strengths of natural sulfur fluxes was deemed desirable. Consequently, simultaneous measurements at three previously-measured sites were undertaken jointly by NOAA, Washington State University Air Resources Laboratory and University of Idaho Department of Chemistry in July and August 1985.

Measurements were based on the use of enclosures that were placed over bare soil, various types of agricultural crops, and naturally occurring vegetative cover, and were flushed with controlled flows of synthetic air or scrubbed ambient air. Concentrations of H_2S, OCS, SO_2, CH_3SH, DMS (CH_3-S-CH_3), CS_2, DES (C_2H_5-S-C_2H_5), MES (CH_3-S-C_2H_5) and DMDS (CH_3-S-S-CH_3) in the enclosures were measured gas chromatographically by the NOAA group using a one-step cryogenic sample enrichment technique in a microbore Teflon capillary sample loop. Concentrations of OCS, DMS, CS_2 and DMDS were measured gas chromatographically by the Washington State group using a two-step cryogenic sample enrichment technique similar to that used by Adams et al. (1). In addition, H_2S was measured by the Washington State group using an impregnated filter - fluorometric quenching technique (2), and total sulfur was measured by the University of Idaho group using a Metal Foil Collection - Flash Vaporization technique (3). Surface and biomass fluxes were related to the concentration measurements by means of the known flush gas flow rate (typically 1.5 Std L/Min), the biomass enclosed (measured by weighing at the end of a suitable measurement period) and the surface area covered.

Although an intercomparison of the results of the various data sets is an important goal of the measurement program, we will discuss here only the details of the NOAA measurements. Preliminary intercomparison has, however, shown that no gross discrepancies exist in the sulfur fluxes measured using the various techniques.

RESULTS

Measurements in the vicinity of Ames, Iowa and Celeryville, Ohio, over bare soil demonstrated a high degree of correlation between the observed fluxes and the enclosure air temperatures. Plots of log (flux) versus temperature showed a linear relationship with correlation coefficients ranging from 0.89 to 0.98. Although in agreement with the results of Adams et al. (1), the observed correlation coefficients were significantly higher than those obtained in the earlier study. The inclusion of various types of actively growing vegetative cover dramatically altered the fluxes of DMS and OCS, the former being significantly increased and the latter decreased. The other significant sulfur flux contributors, H_2S and CS_2, were affected to a much lesser degree by the presence of vegetation. Table 1 summarizes our findings of H_2S, OCS, DMS and CS_2 fluxes from bare soils and soils with different vegetative cover for Ames, Iowa. Similar results were obtained in Celeryville, Ohio, over bare histosol soils and several native "grassy" ground covers. The fluxes measured in this study of approximately 10 ng/S/m^2 min at both Ames, Iowa and Celeryville, Ohio are much smaller than the values of 350 ng S/m^2 min and 130 ng S/m^2 min reported by Adams et al. (1) for the same two sites, respectively.

The results obtained over salt water marshes at Cedar Island, North Carolina were much more temporally and spatially variable and, therefore difficult to summarize. Measurements taken over Spartina Alterniflora in the same location, as were many of the measurements of Adams et al. (1), show obvious tidal effects; a radical diminution of the H_2S flux and a temporary diminution of the DMS flux when the enclosed surface was covered by water. Measurements made over tidal mud flats covered by the species Juncus Romerianus, which appears to comprise more than 95 percent of the tidal marshes in the Cedar Island-Beaufort area, again showed the H_2S flux to have high temporal variability, sometimes changing by more than a factor of 100 in a half-hour period, and frequently surpassing the DMS flux as the dominant species. Although it is difficult to generalize from a few measurements in such a highly variable environment, it appears certain that H_2S and DMS are the dominant species, with OCS, CS_2 and CH_3SH playing a lesser role and DMDS hardly even appearing in quantifiable amounts. The results of our measurements in saline marsh environments are summarized in Table 2. Extreme ranges observed and estimated mean fluxes are listed for H_2S, COS, CH_3SH, DMS and CS_2. No other significant contributors to the natural sulfur flux were observed chromatographically. For comparison, the fluxes measured by Adams et al. (1) and Aneja et al. (4, 5) are also shown.

CONCLUSIONS

The strong correlation of sulfur gas flux with ambient air temperature noted in earlier studies has been corroborated by the present GC measurements at Ames, Iowa and Celeryville, Ohio. Absolute flux values at weighted mean daily temperatures are found, however, to be about a factor of 10, lower than previously reported. Similar measurements carried out over saline marshes at Cedar Island, North Carolina also show a mean flux averaged over tidal cycles, about a factor of 10 lower than previously reported measurements. Concurrent measurements of H_2S and total sulfur fluxes by entirely independent techniques substantiate the lower fluxes reported here.

REFERENCES

1. Adams, D.F., Farwell, S.O., Robinson, E., Pack, M.R., and Bamesberger, W.L., Biogenic sulfur source strengths, Environ. Sci. Tech., 15, 1493-1498, 1981.
2. Natusch, D.F.S., Klonis, H.B., Axelrod, H.D., Teck, R.J., and Lodge, J.P., Sensitive method for measurement of atmospheric hydrogen sulfide, Anal. Chem., 44, 2067-2070, 1972.
3. Kagel, R.A., and Farwell, S.O., Evaluation of different metallic foils for the preconcentration of sulfur-containing gases with subsequent flash desorption/flame photometric detection, Anal. Chem., (submitted, 1985).
4. Aneja, V.P., Overton, J.H., Jr., Cupitt, L.T., Durham, J.L., and Wilson, W.E., Direct measurements of emission rates of some atmospheric biogenic compounds, Tellus, 31, 174-178, 1979.
5. Aneja, V.P., Overton, J.H., Jr., Cupitt, L.T., Durham, J.L., and Wilson, W.E., Carbon disulphide and carbonyl sulphide from biogenic sources and their contributions to the global sulphur cycle, Nature, 282, 493-496, 1979.

TABLE 1. AMES, IOWA, JULY 1985

	Bare Soil Flux $F(T) = AE^{BT}$ (ng S/m^2 min)			Vegetative Cover Flux at 25.5^0		
	A	B	F(25.5)	Oat Grass	Gramma Grass	Soy Plants
H_2S	0.124	0.0643	0.6	3.0±1.0	2.3±0.8	1.0±0.4
COS	0.301	0.0870	2.8	2.0±0.8	1.7±0.6	0.7±0.3
DMS	0.0461	0.115	0.9	6.2±3.4	3.5±1.0	5.3±2.9
CS_2	0.0711	0.0831	0.6	0.6±0.2	0.4±0.2	0.5±0.2
TOTALS			5.0±1.3	11.8±4.4	7.9±2.6	7.5±3.8

TABLE 2. PREDOMINANT NATURAL SOURCES (ng S/m^2 min)
CEDAR ISLAND, NORTH CAROLINA - SALINE MARSHES

	Adams et al			Aneja et al	NOAA, August 1985 Juncus and Spartina Tidally Flooded Marshes	
	10-77	5-78	7-78	7-78	Observed Range	Mean
H_2S	38	38	300	100	3 ---> >10,000	100
COS	4	19	38	80	4 ---> 15	8
CH_3SH			0.6		1 ---> 80	7
DMS	13	76	3000	1500	60 ---> 900	200
CS_2		17	114	400	2 ---> 12	6
TOTALS	55	150	3450	2080		220

DETERMINATION OF NITROGEN EMISSIONS FROM NATURAL SOURCES

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ABSTRACT

The estimated fluxes of NO_x ($NO + NO_2$) from the principal natural sources are presented. According to our present understanding, the largest natural sources of NO_x on the North American continent are lightning and biogenic activity in the soils. However, because of the large uncertainties associated with the estimation of soil emission, improvements in these estimates have been given a high priority. Accordingly, two of the techniques used to measure biogenic NO emissions from the soil have been intercompared. The agreement between the techniques was good. One of these techniques, an enclosure method, has been subsequently used to measure the NO fluxes from the soils at a grassland site in Colorado. The measured NO fluxes compare favorably with NO fluxes measured at a similar site in Australia.

DETERMINATION OF NITROGEN EMISSIONS FROM NATURAL SOURCES

INTRODUCTION

Natural sources of NO_x ($NO + NO_2$) are important, not only because they represent the irreducible minimum for the precursors of the nitrate component of acid precipitation, but also because they dominate the NO_x budget in the upper troposphere, as well as remote regions of the lower troposphere. This budget is important because NO_x controls the photochemical production of ozone and influences the concentration of tropospheric hydroxyl radicals (OH).

Five natural sources of NO_x have been identified: lightning, biogenic processes in soil, oxidation of ammonia, stratospheric injection, and photolytic processes in the oceans. Table 1 lists the emission inventories for the identified sources for global and North American continental averages. The NO_x fluxes contained in Table 1 are based on the estimates of Logan (1983), updated to account for recent developments in this field that have been reported in the current literature. With regards to North America, the emission estimates presented in Table 1 can be summarized as follows: natural NO_x production is dominated by two sources, lightning and soil emissions. Presently, the greatest uncertainty lies in the estimation of soil emissions by NO_x. An improved quantification of NO_x soil fluxes demands the development of improved field methods, intercomparison of new and existing field methods, and extensive field studies. This is the objective of the present study.

INTERCOMPARISON OF METHODS USED TO MEASURE NO FLUXES FROM SOILS

Several techniques have been utilized to measure NO fluxes from the soil, including principally enclosure, gradient, and eddy correlation methods. The enclosure technique measures the NO_x flux from a small, enclosed area of soil surface (typically ~1 m^2), while the gradient and eddy correlation techniques determine the average NO_x fluxes from larger areas (typically ~10^3 m^2). The enclosure method is generally simpler and more convenient than the other two techniques, but it is possible that the fluxes that it is intended to measure are modified due to the disturbance of the soil environment by the presence of the enclosure. Thus far, all the published measurements of NO_x soil (Galbally and Roy, 1978; Johansson and Granat, 1984; Johansson, 1984; Slemr and Seiler, 1984) fluxes have relied on the enclosure technique.

In the present study, a gradient technique for measuring NO fluxes from the soil during nighttime hours has been developed. This technique relies on the measured (NO) gradient with height above the ground to define the vertical mixing and, thus, does not require the simultaneous measurement of micrometeorological parameters. Intercomparison of results from the gradient technique with those from enclosure measurements were made at a site located in Colorado during August and September 1985. The NO fluxes during this period of intercomparison ranged from 0.3 ng N m^{-2} s^{-1} to 25 ng N m^{-2} s^{-1}. Results obtained during this intercomparison are shown in Table 2. The agreement appears to be good over a wide range of NO fluxes.

MEASUREMENTS OF NO_x FLUXES FROM THE SOIL

Following the intercomparison studies, a larger set of flux measurements has been obtained using the enclosure technique. These fluxes were measured from unfertilized, ungrazed, grass-covered soils in a period between 1 August 1985 and 28 October 1985. NO_x fluxes measured during this period using the enclosure technique ranged between 0.03 ng N m^{-2} s^{-1} and 40 ng N m^{-2} s^{-1}. The average flux of NO determined in these studies is in good agreement with the fluxes of NO from ungrazed pastures in Australia measured by Galbally and Roy (1978). The NO flux was found to depend strongly on soil temperature, increasing rapidly with increasing soil temperature. A compilation of these results for data obtained between 5 September 1985 and 28 October 1985 is shown in Table 3. In addition, this sampling period was marked by an extended dry period during the August and October measurement periods. For similar soil temperatures, the NO flux was lowest toward the end of the dry periods and increased sharply with the onset of precipitation. At the end of the extended dry periods, NO_2 is observed to be present in the effluent from the sampling enclosure.

CONCLUSIONS

Present estimates indicate that, pertaining to the North American continent, lightning and biogenic activity in the soil are the largest natural sources of NO_x. However, the estimates of the soil emissions are very uncertain. In the present study, two techniques used to measure biogenic soil emissions, an enclosure technique and a gradient technique, have been intercompared during the nighttime hours. The agreement between the techniques was good. In addition, the enclosure technique has been used to systematically measure the NO_x fluxes at a grassland site and to study the climatological factors that control the NO fluxes. There are no other results published reporting measurements of NO_x fluxes on the North American continent. However, the present results agree with NO fluxes measured at a temperate grassland site in Australia (Galbally and Roy, 1978).

REFERENCES

1. Galbally, J.E., and C.R. Roy, Loss of fixed nitrogen from soils by nitric oxide exhalation, *Nature*, *275*, 734-735, 1978.

2. Johansson, C., Field measurements of emission of nitric oxide from fertilized and unfertilized forest soils in Sweden, *J. Atmos. Chem.*, *1*, 429-442, 1984.

3. Johansson, C., and L. Granat, Emissions of nitric oxide from arable land, *Tellus*, *36B*, 26-37, 1984.

4. Logan, J.A., Nitrogen oxides in the troposphere: global and regional budgets, *J. Geophys. Res.*, *88*, 10785-10807, 1983.

5. Slemr, F., and W. Seiler, Field measurements of NO and NO_2 emissions from fertilized and unfertilized soils, *J. Atmos. Chem.*, *2*, 1-24, 1984.

TABLE 1. ESTIMATES OF NATURAL SOURCES OF NO_x[a]

	Global	North America
Lighting	4	0.3
	(2 - 8)	(0.09 - 0.6)
Soil Emission	8	0.6
	(1 - 16)	(0.08 - 1.5)
Ammonia Oxidation	(0 - 10)	(0 - 0.15)
Stratospheric Injection	0.55	<0.04
	(0.37 - 0.67)	
Oceans	0.2	---
	(0.1 - 1)	---

[a] Units of flux are Tg N yr^{-1}. Estimated range of uncertainty listed in parentheses.

TABLE 2. INTERCOMPARISON OF BOX AND GRADIENT MEASUREMENTS OF NO FLUX[a]

Date	Box	Gradient
8/24	0.35	0.88
8/25	0.28	0.20
8/26	0.83	0.81
8/27	0.83	1.49
8/30	0.37	0.21
8/31	0.37	0.16
9/01	20.0	17.0

[a] The result gives the value (or average value) of the NO flux determined during the nighttime hours of each of these dates. Units of flux are Ng N m^{-2} s^{-1}.

TABLE 3. THE AVERAGE FLUX OF NO MEASURED AS A FUNCTION OF SOIL TEMPERATURE OVER 5° INCREMENTS OF SOIL TEMPERATURE[a]

Soil Temperature Range (°C)	Average Flux NO (ng N m^{-2} s^{-1})
0 - 5	0.076
	(0.028 - 0.16)
5 - 10	0.34
	(0.075 - 1.4)
10 - 15	0.94
	(0.20 - 3.9)
15 - 20	2.5
	(0.72 - 10)
20 - 25	4.5
	(2.4 - 7.3)
25 - 30	3.9
	(2.9 - 8.1)

[a] The range of measured fluxes for each temperature increment is listed in parentheses.

ALKALINE AEROSOLS EMISSIONS

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ABSTRACT

Alkaline compounds contained in aerosols emitted from open (non-combustion) sources may have the potential to neutralize acid precipitation. This potential depends on the quantity of aerosol mass emitted, its transport, fraction of alkaline compounds in the total aerosol mass, solubility of the alkaline compounds, and neutralization potential. It is the mission of the Alkaline Aerosols Project to estimate the total neutralizing alkaline aerosol fluxes by evaluating the above factors.

Provisional estimates of alkaline aerosol fluxes showed the following:

(1) That the two most important sources (unpaved road dust and wind erosion) dominate all other sources.

(2) That methods to estimate total alkaline aerosol emissions give total fluxes that are larger than observed deposition rates. This and other inconsistencies of the estimates show that strong efforts are needed to develop new methods to estimate emissions of alkaline aerosol.

(3) Knowledge is lacking on the fraction of suspended emitted aerosols. Knowledge is also lacking on solubility and neutralization potential of the aerosol.

Research efforts have been planned to estimate dust emissions from wind erosion, dust devils, and unpaved roads. Solubility and neutralization potential of aerosols produced by these sources will be determined for several experiments.

OPEN SOURCES OF ALKALINE AEROSOLS

INTRODUCTION

Alkaline compounds carried in aerosols emitted from open sources may have the potential to neutralize acid rain. To determine the importance of open sources of aerosols for neutralizing precipitation, a systematic approach was laid out to estimate the flux of alkaline aerosol having sufficient residence time in the atmosphere to interact with precipitation systems. After a general framework of the estimation was laid out, existing data and models were used to give a provisional estimate of alkaline aerosol emissions. Additional work was undertaken to determine solubility and neutralization potential of the aerosols. The estimate for quantity of emitted soluble alkaline aerosols was examined to find shortcomings and to plan a more thorough estimation method.

DISCUSSION

Emission fluxes of alkaline aerosols (F) that have potential to neutralize acid rain are expressed as follows:

$$F = \frac{E\ A'\ Cf\ SX\ N}{A} \qquad (1)$$

E = Emission factor [mass/(area yr)]

A' = Area of emission

C = Elemental abundance of parent material (fraction of mass)

f = Fractionation of aerosol composition to parent material composition

S = Fraction of aerosol mass suspended

X = Solubility of suspended aerosol

N = Neutralization factor (fraction)

A = Total area in question.

An initial evaluation of eq. (1) was made state-by-state. The values for EA' were those reported by Evans and Cooper (1980). Relative abundance of the alkaline compounds was assumed to be the same in the aerosols as in the parent material (fractionation of unity). Solubility for alkaline elements was taken to be that derived from two soluble/insoluble data sets; daily bulk data from St. Louis and 1 year of weekly sampling data for wet side precipitation from Glen Ellyn, Illinois (Gatz, et al., 1984). The composition of the parent material was taken to be the median value of the state soil chemical survey data (Boerngen and Shacklette, 1981) for soil erosion and soil tilling and to be a combination of median state soil chemical composition and chemical composition of other road surface materials (see Barnard, et al., 1985) for unpaved roads. The fraction of aerosol mass suspended was 0.4 based on the fraction of mass smaller than 10 μm and published aerosol size distributions. Neutralization potential (N) was set to one even though this can only be an upper limit.

Results of the provisional evaluation of eq. (1) are shown for the 31 eastern states in Table 1 (after Gatz, et al., 1985). The results show the dominance of the two most important open sources (unpaved roads and wind erosion) over conventional sources for all the alkaline elements but sodium, which has little neutralizing potential.

Table 1. (after Gatz, et al., 1985). Total mass and estimated cation emissions from major open and conventional sources in the 31 "eastern" states. Emission estimates for open sources are limited to soluble materials < 10 μm; these limitations do not apply to the estimates of emissions from conventional sources.

	Thousands of tonnes/yr (1980)*				
Source category	Total mass	Na	Mg	K	Ca
OPEN**					
Wind erosion	37,500	107	47.4	123	109
Tilling	7,420	18.9	8.28	23.7	18.7
Unpaved roads	117,000	172	531	231	4,110
Sum	162,000	297	587	378	4,240
CONVENTIONAL***	8,410	109	119	71.6	274

* Rounded to three significant figures.

** Total mass emissions are from Evans and Cooper (1980); element emissions are based on the work of Stensland et al. (1985).

*** Sum of separate emissions estimated for the following categories: Fuel combustion, industrial processes, solid wate disposal, transportation, and miscellaneous (less unpaved roads). Total mass emissions are from U.S. EPA (1982); element abundances used to calculate emissions are from Gatz et al. (1985).

Comparisons on a state-by-state basis gave total mass fluxes that are much larger than downward fluxes calculated from NADP/NTN data. This poor agreement pointed to overestimation by the provisional evaluation of eq. (1) and pointed to needed research especially emission factors (E), fractionation factors (f), solubility (X), chemical abundance of roads (c), and neutralization potential (N).

To correct for this overestimation and to improve our knowledge of alkaline aerosols our plans for research include:

1). Development of new wind erosion emission equation. The equation will be physically based and will rely on results of past research and new research.

2). Development of dust devil emission estimations. These estimates will be based on aircraft based dust devil sampling and meteorological classification/generalization to the American Southwest.

3). Development of a new emission equation for unpaved roads. This will be based on new experimentation and theory of dust emissions by vehicles on unpaved roads.

4). Experimentally determined fractionation factors. These factors will be developed by examining ratios of relative composition of emitted aerosols to parent material for both unpaved road emissions and wind erosion.

5). Experimentally determined solubility factors. This effort will be for alkaline aerosols material collected in experiments on road emissions and wind erosion.

6). Chemical survey of unpaved roads in the U.S. This chemical survey will fill a gap in knowledge on the parent materials of U.S. unpaved roads.

7). Experimentally determined neutralization potentials. This work is necessary because alkaline cations are not always associated with anions such that H ions are readily replaced. Composition of the alkaline aerosols must be carefully studied.

CONCLUSIONS

The present state-of-the-art in estimating alkaline aerosol emissions having potential to neutralize acid rain gives numbers which are larger than NADP/NTN observed deposition by roughly a factor 2-3. The research needed to improve the estimates will include development of emission equations for wind erosion and unpaved roads. Research into solubility, neutralization potential and fractionation factors will be incorporated into the new estimations. Chemical surveys of the national unpaved roads will also be executed.

REFERENCES

Barnard, W. R., G. J. Stensland, and D. F. Gatz, 1985: Alkaline materials flux from unpaved roads: Source strength, chemistry and potential for acid rain neturalization. Submitted to Water, Air, and Soil Pollution.

Boerngen, J. G., and Shacklette, H. T., 1981: Chemical analysis of soil and other surficial materials of the conterminous United States. Open File Report 81-197, U.S. Geological Survey, U.S. Dept. of Interior, Denver, Colo.

Evans, J. S., and Cooper, D. W., 1980: An inventory of particulate emissions from open sources. J. Air Pollut. Control Assoc., 30, 1298.

Gatz, D. F., W. R. Barnard, and G. J. Stensland, 1985: The role of alkaline materials in precipitation chemistry: A review of the issues. Submitted to Water, Air and Soil Pollution.

Gatz, D. F., Stensland, G. J. Miller, M. V., and Chu, L.-C., 1984: Alkaline aerosols: An initial investigation of their role in determining precipitation acidity. Final Rept. NSF Grant ATM 77-24294. Illinois State Water Survey Contract Report No. 343.

METHODOLOGY FOR ESTIMATING NATURAL HYDROCARBON EMISSIONS

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ABSTRACT

An emission inventory system for biogenic sources of hydrocarbons has been developed. It is based on modifications of the classic formula:

$$\text{Emissions} = \Sigma \text{ Biomass} \star \text{Area} \star \text{Emission Factor}$$

It accomodates multiple sources with emission factors dependent on season, temperature and solar intensity. It provides emissions on a latitude longitude based grid system aggregated by plant specie, hydrocarbon compound, study area (county, state, etc.), or time interval. The emission calculations from this inventory have better spatial, source, temporal and hydrocarbon compound resolution than prior work has provided.

INTRODUCTION

Part of the evaluation of the Regional Acid Deposition Modeling effort will include natural source contributions and effects. The new chemistry model has been extended to incorporate biogenic hydrocarbons in the simulation. The use of kinetic reactions employing natural sources of hydrocarbons in air and water requires definition of their emissions. Such an emission inventory should recognize production of biogenic hydrocarbons from:

- the foliage of trees, shrubs, and crops
- decaying leaf litter
- grasses in urban areas, pastures, and grasslands
- decaying vegetation in rivers, lakes, marshes, and oceans.

Methane, monoterpenes, and isoprene are the dominant hydrocarbon compounds produced by biogenic sources.

A systematic methodology was needed for the estimation of biogenic hydrocarbon emissions. Such an inventory should have the following capabilities:

- Generate hourly gridded emissions
  - (a) for a variable number of hydrocarbon species;
  - (b) from a variable number of plant species, leaf litter, and surface waters;
  - (c) for any latitude/longitude-based grid system.
- Apply temporal variations to emission factors on a diurnal and/or seasonal basis.
- Adjust emission factors for variations in temperature and solar radiation.
- Allow for future modification of emission factors to incorporate submodel calculations, including leaf temperature, foliar density, soil types, moisture availability, and disease prevalence.
- Provide summary reports of emissions by grid cell, county, U.S. Forest Service (USFS) region, and state.

Such a system has been developed for the Northeastern United States and offers the potential for meeting one of the requirements of the Regional Acid Deposition Model.

DISCUSSION

Biogenic hydrocarbon emissions are a function of emission rate and biomass within a given area. An emission inventory for them includes of building files of descriptive data on land use, emitter density, and emission factors. It also includes algorithms implementing the relations between the data elements of these descriptive files.

Area categorization has been primarily derived from the USGS Land Use and Land Cover map series supplemented as needed from other satellite and aerial sources. Coverage areas for the following land use classifications were estimated to the nearest five percent for each grid cell:

- -urban
- -agricultural
- -rangeland
- -deciduous forest
- -coniferous forest
- -mixed forest (including forested wetland)
- -water
- -barren land
- -nonforested wetland
- -mixed agricultural and rangeland
- -rocky open areas occupied by low growing shrubs and lichen (not including tundra)

Mixed forest was split between the coniferous and deciduous classifications, with data from the U. S. Forest Service inventories used to allocate area to individual species. The canopy area is a function of the diameter at breast height (dbh), which is divided into ten intervals or classes. The area for one tree in a dbh class multiplied by the number of trees in the class and summed across classes gives the total area for a specie. A mix ratio of the specie area to the total area summed across all species of the same forest type, deciduous or coniferous, was used to allocate the proportion of the corresponding forest type area to that specie. Agricultural land was apportioned among crops based on the 1978 Census of Agriculture as aggregated to the county level by the Oak Ridge National Laboratory. Several state atlases were used to resolve the split between fresh and salt water, especially in estuaries. A file of the overlap between grid cells and study unit (county, forest region, etc.) was used to partition classification areas among the grid cells. It is used in reverse to summarize emissions at the study unit level.

Most of the hydrocarbons emitted by vegetation are released by the foliage. The branches, twigs, bole, and root emissions are negligible in comparison. Foliage weight can be estimated from algorithms which relate foliage weight to tree diameters. Tree diameter inventories are compiled by the U. S. Forest Service for each state. Emission factors for surface waters, leaf litter, and some crops are area dependent and do not require a foliar weight estimate.

The emission factor is expressed as moles or micrograms of hydrocarbon emitted over a time interval per unit of land area or dry foliar weight. The identity of the hydrocarbons emitted is reported as isoprene, monoterpene, and/or unspecified nonmethane hydrocarbon. When emission factors were unavailable for a particular vegetative type, an emission factor from a biologically similar vegetation type was used. These emission factors have several possible sources of error:

- Measurement error due to the experimental method employed disturbing the natural emission process.
- Inappropriate substitution of emission factors due to unavailable data.
- Emission factors may vary with environmental parameters including geographic location, soil conditions, and elevation.

Because emission factors are known to vary with light and temperature, the system accounts for seasonal changes in solar radiation and surface temperature. It is assumed that the leaves of all plants are exposed to full sun and the same surface temperature. Because there may be large variations in solar radiation and surface temperature, hourly estimates are made of these variables. Solar radiation is derived from formulas that give the solar zenith angle as a function of latitude, longitude, date, and time of day. The system does not currently account for attenuation due to clouds and aerosols, but provision has been made for this variable in future versions of the system. Thus, the emissions generated by the system are maximum rates with respect to the effects of sunlight. Surface temperatures from the National Weather Service are transformed to average grid cell temperatures by an objective mapping technique for mesoscale processes based on numerical filtering. All of the emission factors have been standardized to 30°C. It has been assumed that all monoterpene emitters and nonmethane hydrocarbon emitters (other than isoprene) respond to temperature in a manner similar to monoterpene emissions from slash pine. Likewise all isoprene emitters have been assumed to respond to changes in light and temperature as does isoprene from live oak.

CONCLUSIONS

An emission inventory system has been developed to meet the requirements of the new kinetic reactions employing natural sources of hydrocarbons in air and water. It provides much better resolution than prior inventories especially in terms of spatial and temporal variation. It is open ended allowing inclusion of different areas, sources, emission factors and temporial variations. It will provide insight into the relative contribution of a few species to the total emissions in an area. A systematic, detailed inventory increases the confidence in the emissions since variances can be reasonably calculated. Future research on emission factor development, module extensions, etc. can be guided by the knowledge gained from the system.

SESSION 5: RELATED EMISSION INVENTORY DEVELOPMENT ACTIVITIES

Chairman: J. David Mobley
Air and Energy Engineering Research Laboratory (MD-61)
U. S. Environmental Protection Agency
Research Triangle Park, NC 27711

SUMMARY OF A 1982 NATIONAL EMISSIONS INVENTORY FOR REGIONAL MODEL DEVELOPMENT

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Presented at:

Second Annual Acid Deposition Emission Inventory Symposium

Charleston, South Caroline

November 13-14, 1985

SUMMARY OF A 1982 NATIONAL EMISSIONS INVENTORY FOR REGIONAL MODEL DEVELOPMENT

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ABSTRACT

None of the regional and national emissions inventories available in 1982 were suitable for regional deposition or air quality modeling. Therefore, procedures were developed and applied to produce a suitable 1982 inventory for the United States and southeastern Canada. The inventory includes total emitted particulate matter (TEP), alkaline TEP, SO_2, primary SO_4, NO, NO_2, NH_3, and hydrocarbons in nine photochemical reactivity classes. Point source geographic locations are resolved to ±100 meters, and stack data are available for calculating emission injection height. Area source emissions are available by county or within 1/4° longitude by 1/6° latitude elements of a grid system. The inventory contains annual, seasonal, weekday, weekend, and 3-hourly average emission rates. United States emissions of TEP, SO_2, NO_x and HC in the inventory are 1,698, 26.5, 23.8, and 20.8 million tons per year, respectively. The point source data are from various years; about one-half of the SO_2 and NO_x emissions pertain to 1982. Older data for some states are currently being updated. This inventory is currently being compared with the 1980 National Acid Precipitation Assessment Program (NAPAP) inventory by source category and state to identify and evaluate differences. Procedures used to develop them are also being compared to evaluate consistency between the inventories and improve inventory development procedures in general.

SUMMARY OF A 1982 NATIONAL EMISSIONS INVENTORY FOR REGIONAL MODEL DEVELOPMENT

INTRODUCTION

None of the regional and national emissions inventories that were available in 1982[1,2,3,4,5] were suitable for regional modeling applications throughout the United States and eastern Canada. They were limited with respect to geographic coverage, data concurrency, chemical substances, or spatial resolution, or specification of uncertainties. To improve upon the situation, the Electric Power Research Institute (EPRI) sponsored the development and application of procedures to compile a 1982 emissions inventory for the contiguous United States and southeastern Canada[6,7].

The inventory includes total emitted particulate matter (TEP), alkaline TEP, SO_2, primary SO_4, NO, NO_2, NH_3, and hydrocarbons (HC) in nine photochemical reactivity classes. Point sources geographic locations are nominally resolved to ±100 meters. Stack heights, diameters, flow rates, and exit temperatures are included for calculating emission injection height. Area source emission rates are available by county. Factors to allocate these county-wide data to elements of a 1/4° longitude by 1/6° latitude grid system are also available. The inventory contains annual, seasonal, weekday, weekend, and diurnal (3-hour resolution) average emission rates.

DISCUSSION

Annual emissions from point and area sources are in Table 1. Almost all TEP emissions are from wind erosion and agricultural tilling area sources. Point sources emit over 90% of the SO_2 in the country. Point and area sources contribute equally to NO_x emissions, and area sources account for 70% of HC emissions.

Although the nominal time period of the inventory is 1982, it contains point source data from several years. The national distribution of annual point source emissions by year-of-record is in Table 2. About one-half of the SO_2 and NO_x emissions and 30% and 27% of the TEP and HC emissions are for 1982. Less than half of the point source emissions for any material pertain to years before 1981.

Most point source SO_2 and NO_x emissions are from a relatively small number of large sources. Almost 80% of point source SO_2 emissions are from 406 facilities that each emit more than 10,000 tons per year, and about 70% of point source NO_x emissions are from 440 facilities that each emit more than 5,000 tons per year. The 1,289 largest emitters account for about 73% of point source TEP emissions, and the 784 largest emitters account for about 75% of point source HC emissions.

Annual emissions by source category are shown in Table 3. Almost all of the TEP is emitted by fugitive area sources, particularly wind erosion and agricultural tilling. Emissions from unpaved roads are also substantial. Industrial processes are the largest point source contributors to national TEP emissions, and residential fuel use is the largest combustion source. TEP emissions from mining, which could be important in some parts of the country, are not in the inventory.

Fossil-fueled electric utility boilers account for 66% of SO_2 emissions. Industrial boilers and industrial processes each contribute 14%. The remaining 6% is from other fuel combustion.

Electric utility and industrial boilers account for 53% of national NO_x emissions, and motor vehicles contribute 30%. Industrial processes, which include refinery and chemical plant process heaters, contribute 9%.

Industrial processes, motor vehicles, miscellaneous solvent uses, and residential fuel combustion are the largest HC emitters. Most industrial process emissions are from storage tanks and solvent use. Wood combustion accounts for most HC emissions from residential fuel use.

Annual NH_3 and alkaline TEP emissions are listed by source category in Table 4. Emissions from bacterial action in the ground and from livestock account for most of the NH_3 in the inventory. However, emissions from fertilizer use are not included. This category may be significant. Wind erosion and agricultural tilling emit most of the alkaline TEP.

Tables 5 and 6 show comparisons of total United States SO_2 and NO_x emissions with Version 3.0 of the 1980 National Acid Precipitation Assessment Program (NAPAP) inventory[8]. Although these national totals agree within 5%, some of the procedures and data sources used to develop the inventories were different. In particular, electric utility data in NAPAP were derived from forms filed by utilities with the Department of Energy, while data in the EPRI inventory came from the National Emissions Data System[1], state inventories, and some plant operators. These and other differences may have produced significant differences in emission rates from some source categories in some regions of the country. It is important that such differences be identified and evaluated, since they could affect results of regional modeling applications.

Procedures used to calculate primary SO_4, NO, NO_2, and HC reactivity class emissions also differed between the inventories, as did procedures used to calculate seasonal, day-of-week, and diurnal average emission rates. The effects of these differences must also be evaluated.

CONCLUSIONS

The data in the inventory indicate that area sources, particularly wind erosion and agricultural tilling, dominate annual United States TEP and alkaline TEP emissions. Point sources, principally electric utility boilers, emit most of the SO_2 in the United States. Point and area sources emit nearly equal amounts of NO_x. Motor vehicles, industrial processes, miscellaneous solvent uses, and residential combustion are the major sources of HC. Animals and bacterial action in the ground are the major sources of NH_3.

The inventory does not currently include fugitive TEP emissions from surface mining, which could be significant in some parts of the country. NH_3 emissions from fertilizer use also are not included. These sources will be added to the inventory in the future.

A major fraction of national point source emissions in the inventory pertain to time periods before 1982. Work is under way to update some of the older values to improve data concurrency.

Although total SO_2 and NO_x emission rates for the United States are almost the same in the EPRI and 1980 NAPAP inventories, some data sources and procedures used to develop these inventories were different. These differences may have caused emissions from some sources to differ significantly in some parts of the country. Therefore, these inventories are currently being compared by source category and state to identify and evaluate differences. Procedures used to develop the two inventories are also being compared to evaluate the consistency between the inventories and, hopefully, improve inventory development procedures in general.

Acknowledgement. This work was supported under Electric Power Research Institute Contract RP1630-23.

REFERENCES

1. AEROS Manual Series; Volume II: AEROS User's Manual, USEPA Research Triangle Park, NC, EPA-450/2-76-029, December 1976.

2. H. A. Klemm and R. J. Brennan, *Emissions Inventory for the SURE Region*, GCA/Technology Division for the Electric Power Research Institute (EPRI), EA-1913, April 1981.

3. C. M. Benkovitz, "Compilation of an Inventory of Anthropogenic Emissions in the United States and Canada." *Atmos. Environ*. 16 (6): 1551 (1982).

4. F. M. Sellars, et al., *Northeast Corridor Regional Modeling Project Annual Emission Inventory Compilation and Formatting, Volumes I through XVIII*, EPA-450/4-82-013a-r, August 1982-July 1983.

5. *Emissions, Costs, and Engineering Assessments*, Work Group 3B, U.S./Canada Memorandum of Intent on Transboundary Air Pollution, Junr 1982.

6. S. L. Heisler, "Emission Inventory Requirements for Deposition and Regional Air Quality Model Development: A Summary." In: *Proceedings: First Annual Aicd Deposition Emissions Inventory Symposium*, Research Triangle Park, NC, EPA-600/9-85-015, May 1985.

7. S. L. Heisler, et al., "The EPRI 1982 Air Pollutant Emission Inventory." Presented at the 78th Annual Meeting of the Air Pollution Control Association, Paper 85-4.3, Detroit, MI, June 16-21, 1985.

8. D. A. Toothman, et al., *Status Report on the Development of the NAPAP Emission Inventory for the 1980 Base Year and Summary of Preliminary Data*, Research Triangle Park, NC, EPA-600/7-84-091, December 1984.

3. C. M. Benkovitz, "Compilation of an Inventory of Anthropogenic Emissions in the United States and Canada." Atmos. Environ. 16 (6): 1551 (1982).

4. F. M. Sellars, et al., Northeast Corridor Regional Modeling Project Annual Emission Inventory Compilation and Formatting, Volumes I through XVIII, EPA-450/4-82-013a-r, August 1982-July 1983.

5. Emissions, Costs, and Engineering Assessments, Work Group 3B, U.S./Canada Memorandum of Intent on Transboundary Air Pollution, Junr 1982.

6. S. L. Heisler, "Emission Inventory Requirements for Deposition and Regional Air Quality Model Development: A Summary." In: Proceedings: First Annual Aicd Deposition Emissions Inventory Symposium, Research Triangle Park, NC, EPA-600/9-85-015, May 1985.

7. S. L. Heisler, et al., "The EPRI 1982 Air Pollutant Emission Inventory." Presented at the 78th Annual Meeting of the Air Pollution Control Association, Paper 85-4.3, Detroit, MI, June 16-21, 1985.

8. D. A. Toothman, et al., Status Report on the Development of the NAPAP Emission Inventory for the 1980 Base Year and Summary of Preliminary Data, Research Triangle Park, NC, EPA-600/7-84-091, December 1984.

Table 1. ANNUAL UNITED STATES POINT AND AREA SOURCE EMISSIONS

	Emissions (million tons/year)			
	TEP	SO_2	NO_x	HC
Point Sources	5.1	24.4	11.9	5.9
Area Sources	1693	2.1	11.9	15.9
Total	1698	26.5	23.8	20.8

Table 2. DISTRIBUTION OF INVENTORIED UNITED STATES POINT SOURCE SMISSIONS BY YEAR-OF-RECORD

Emissions (percent of total)
(million tons/year)

	Before 1980	1980	1981	1982	After 1982
TSP	1.4 (27%)	0.7 (14%)	0.9 (17%)	1.5 (30%)	0.6 (12%)
SO_2	1.8 (7%)	2.5 (10%)	5.5 (23%)	13.0 (53%)	1.5 (6%)
NO_x	1.3 (11%)	1.3 (11%)	2.8 (24%)	5.8 (48%)	0.8 (6%)
HC	2.2 (37%)	0.9 (15%)	0.6 (10%)	1.6 (27%)	0.6 (10%)

Table 3. ANNUAL UNITED STATES EMISSIONS BY SOURCE CATEGORY

Source Category	Emissions (thousand tons/year) TEP	SO_2	NO_x	HC
Stationary Source Fuel Combustion				
Electric Utilities	800	17,380	7,920	59
Industrial Boilers	588	3,699	4,138	1,039
Commercial/Institutional	115	610	354	15
Residential	1,011	177	404	2,089
Subtotal	2,514	21,866	12,816	3,202
Industrial Processes				
Non-Ferrous Smelters	184	1,092	47	16
Other Processes	3,457	2,690	1,994	5,529
Subtotal	3,641	3,782	2,041	5,545
Solid Waste Disposal	418	44	176	677
Transportation				
Motor Vehicles	129,519[a]	365	6,869	6,195
Airplanes	82[b]	12	109	161
Vessels	26	172	141	427
Railroads	49	112	725	176
Other Off-Highway	59	114	827	517
Subtotal	129,735	775	3,671	7,476
Miscellaneous				
Wind Erosion	956,300	0	0	0
Agricultural Tilling	595,000	0	0	0
Construction	9,432	0	0	0
Cattle Feedlots	440	0	0	0
Forest Fires	288	3	68	325
Other Burning	560	3	102	417
Gasoline Marketing	0	0	0	960
Miscellaneous Solvents	0	0	0	3,157
Subtotal	1,562,020	6	170	4,859
Total	1,698,328	26,473	23,874	21,759

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[a] Includes entrained particulate matter from road surfaces.

[b] Includes entrained particulate matter from unpaved air strips.

Table 4. ANNUAL UNITED STATES NH_3 AND ALKALINE TEP EMISSIONS

	Emissions (thousand tons/year)	
	NH_3	Alkaline TEP[a]
Fuel Combustion	774	0
Industrial Processes	23	0
Unpaved Roads	0	3,972
Paved Roads	0	111
Construction	0	300
Wind Erosion	0	46,960
Agricultural Tilling	0	18,880
Cattle Feedlots	1,189	168
Other Livestock	4,900	0
Land Surfaces	5,103	0
Human Beings and Pets	266	0
Total	12,255	70,391

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[a] Expressed as equivalent calcium carbonate.

Table 5. COMPARISON OF ANNUAL SO_2 EMISSIONS WITH THE 1980 NAPAP INVENTORY

	Emissions (million tons/year)	
	NAPAP[a]	EPRI
Electric Utilities	17.3	17.4
Non-Utility Combustion	4.6	4.5
Non-Ferrous Smelters	1.2	1.1
Transportation	0.9	0.8
Other Sources	3.1	2.7
Total	27.1	26.5

Table 6. COMPARISON OF ANNUAL NO_x EMISSIONS WITH THE 1980 NAPAP INVENTORY

	Emissions (million tons/year)	
	NAPAP[a]	EPRI
Electric Utilities	8.1	7.9
Non-Utility Combustion	5.2	4.9
Non-Ferrous Smelters	0	0
Transportation	9.1	8.7
Other Sources	1.3	1.3
Total	23.7	22.8

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[a] National Acid precipitation Assessment Program 1980 inventory, Version 3.0[8].

GENERATION OF TEMPORALLY-RESOLVED EMISSIONS INVENTORIES FOR CANADA

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Second Annual Acid Deposition Emission Inventory Symposium
Charleston, South Carolina
November 12-14, 1985

ABSTRACT

Processing of North American anthropogenic and natural source emissions for the Canadian long-range transport model (ADOM) involves the generation of an hourly speciated emissions file on the 127 km Eulerian grid. Some anthropogenic and most natural emissions are strongly dependent on local climate and hourly meteorology. The use of generally derived or average temporal factors for these sources can lead to inconsistencies between between prevailing meteorology and the estimated emission rates used in evaluating the ADOM model. This paper describes a system for preprocessing of anthropogenic and natural source emissions using the ADOM database of hourly gridded meteorological data and other temporally variable parameters to provide an hourly emissions file which avoids some of the inconsistencies inherent in the use of average temporal factors.

1. INTRODUCTION

A Eulerian long-range transport model is presently being developed by the Ontario Ministry of the Environment, with Environment Canada and the Federal Republic of Germany as co-sponsors. This model, known as the ADOM model, includes a model of the atmospheric chemistry which requires a detailed knowledge of the reactant mix in each 127 x 127 km grid cell. This mix is partly dependent on the spatial and temporal distribution of speciated emissions. While presently available emissions inventories provide reasonably good spatial resolution of the annual average emissions, temporal factors are generally based on typical seasonal and diurnal variations rather than year specific or hourly meteorological data. The use of typical temporal profiles for climatologically and meteorologically dependent sources such as open anthropogenic and natural sources can lead to inconsistencies between the prevailing hourly meteorology and the estimated emission rate. Since under some circumstances, open sources and other meteorologically dependent emissions can overwhelm their better defined process-related counterparts, it is important for purposes of model evaluation to provide a more realistic temporal resolution of these emissions. Part of the database for the ADOM model comprises hourly meteorological data as well as soil moisture and temperature, gridded to the 127 km Eulerian grid for the subject period. The availablity of this database, facilitates the more realistic estimation of meteorologically forced emissions.

This paper describes an emissions preprocessor for anthropogenic and natural sources which provides a first level of refinement in preparing an hourly resolved emissions file and which avoids many of the inconsistencies inherent in using typical temporal factor profiles. This preprocessor will have as its first application, the preparation of an emissions file for the 1980 PEPE/NEROS experimental period. Details of the methodology used to derive seasonal, day-of-week and diurnal temporal factors for anthropogenic area sources and natural sources are given in References (1) and (2).

2. DISCUSSION

2.1 Emissions Processing System for the ADOM

Figure 1 shows the general steps taken in processing the (1980) Canadian and U.S. point and area source emissions. The Canadian emissions file contains annual average 1980 anthropogenic emissions for area and minor point sources together with temporal factors for the PEPE/NEROS period for a selection of major point sources. The area source emissions are provided on a 127 x 127 km gridded basis. The U.S. anthropogenic emissions inventory is on a county basis and includes some temporal factor information as part of the file. The system provides a facility to allocate the U.S. emissions to the ADOM grid, reformat and merge them with the Canadian data to provide a single emissions file. This merged file is then processed through the emissions preprocessor which applies the temporal factors to process-oriented emissions and which uses appropriate climatological and meteorological data to generate temporally and spatially resolved open anthropogenic and natural source emissions. The output from the system is a file of gridded hourly emission rates for a specified period. While the system is intended to process both Canadian and U.S. sources, the climatological data and other emission parameter data for open and natural sources as well as other meteorologically dependent sources have so far only been prepared for Canada for the year 1980.

2.2 Temporal Emissions Preprocessor

Figure 2 shows the subsystem for generation of temporally resolved emissions. For purposes of this preprocessor, emissions are divided into the following three classes:

(i) Production, Consumption and Process Related Emissions

For sources in this class, the preprocessor function is simply to apply prescribed seasonal, day-of-week and diurnal temporal factors to the annual average emission rates. Some sectors in this class do have a climatic component (for example, gasoline and diesel marketing, motor vehicle and residential heating emissions). The gridded annual average emissions in A are processed by G using seasonal, day-of-week and diurnal factors contained in file B. Seasonal factors are based on 1980 quarterly and monthly production, consumption or other statistics available from Statistics Canada. These statistics give an estimate of the province-wide monthly or quarterly relative activity variations. The annual average emissions are available for Canada on the 127 km grid and the seasonal activity profile and therefore the seasonal temporal factors are assumed to be uniform throughout the province. Where climate dependence is significant (Table 1) the normalized seasonal profiles of the relevant climate variable/s have been estimated for some thirty-five climate zones of approximately 4° in latitude width as shown in Figure (3) from Reference (2). These zonal factors which reflect the intra-province variation of climate extremes are used to weight the activity related temporal factors to resolve the spatial variation of seasonal temporal factors in each province. These spatially resolved seasonal factors are contained in file B. Where possible, 1980 climate data have been used; heating degree day distribution for example. Figures 4 and 5 from Reference (1) are examples of the computed latitude variation of seasonal temporal factors for diesel and gasoline marketing and gasoline powered motor vehicles.

Specific data on day-of-week or diurnal activity profiles were not available and these temporal factors are extracted from other studies as well as from a limited survey of industry practice.

(ii) Open Anthropogenic Sources

Sources in this class (Table 1) are mainly contributors to suspended particulate bearing alkali and alkaline earth metals as well as iron and manganese. The alkaline material is important for pH dependent wet chemical reactions while the Fe and Mn ions catalyse wet SO_2 oxidation. Open source particulate emissions are strongly dependent on prevailing meteorology, vegetative and snow cover, as well as silt and moisture content. Emissions in this class are computed in module E using the hourly gridded meteorological data in C. In some cases an emissions algorithm is used to estimate the grid average rate, for others only a likelihood weighting factor for certain activities can be assessed on the basis of meteorology (for example spraying activities, slash burning). Some emissions which are primarily determined by non-meteorological factors such as road dust are suppressed during and subsequent to precipitation events. Again, climate related parameters in the emission algorithm have been estimated on a 4^o latitude basis while parameters such as soil type and area of agricultural land have been gridded to the 127 km resolution.

(iii) Natural Sources

Apart from time-stationary factors such as vegetation and soil type, natural source emissions are entirely determined by climatic and meteorological factors. While some annual average emission estimates are available for natural sources, these are used where only seasonal variation is resolved, for all other natural sources, the hourly emission rates are determined using an emission algorithm together with zonal climate data and hourly gridded meteorological data (Table 2).

3. CONCLUSIONS

In order to resolve natural and some anthropogenic emission rates to the spatial and temporal scales required by the ADOM Eulerian long-range transport model, it is necessary to consider the impact of spatial climate variability as well as hourly meteorology on the temporal variation of emission rates. Using the hourly gridded meteorological fields and other data from the ADOM database it is possible to prepare an hourly emissions record which refines the emissions estimates by including these effects.

REFERENCES

1. MEP Company and Ontario Research Foundation, "Temporal Factors for 1980 National Anthropogenic Area Source Emissions", Report prepared for Environment Canada, July 1985.

2. Environmental Applications Group Ltd., "Gridded Natural Emissions Inventory and Temporal Algorithms", Report prepared for Environment Canada, May 1985.

Table 1

Climatologically and Meteorologically Dependent Anthropogenic Area Sources

Source	Parameters
Gasoline Powered Motor Vehicles	Seasonal mean e_f (T)
Gasoline and Diesel Marketing	Monthly mean e_f (T)
Residential Heating	Seasonal degree days
Paved and Unpaved Roads	Precipitation
Pesticide Application	Wind, Precipitation
Slash Burning	Wind, Precipitation
Tailings Piles	Wind, Seasonal PE
Landfill Sites	Growing degree days
Tilling	Precipitation

Table 2

Natural Sources

Source	Seasonal	Diurnal	Species	Parameters
Marine Salt		X	Ca, Mg, K, SO_4, Fe, Mn	wind, ice cover
Soils		X	NH_3, VOC, NOX, $S(H_2S)$	soil temperature, air temperature, snow cover
Vegetation		X	VOC	vegetative state, air temperature
Soil Erosion		X	K, Mg, Ca, Fe, Mn, particulate	wind, snow cover, vegetative state, soil moisture
Lightning		X	NOX	climatological
Animal Wastes	X		NH_3, VOC	climatological
Forest Fires	X		SO_2, CO, NOX, VOC, Ca, Mg, K, SO_4, particulate	climatological
Marines Waters	X		VOC, CO	ice cover

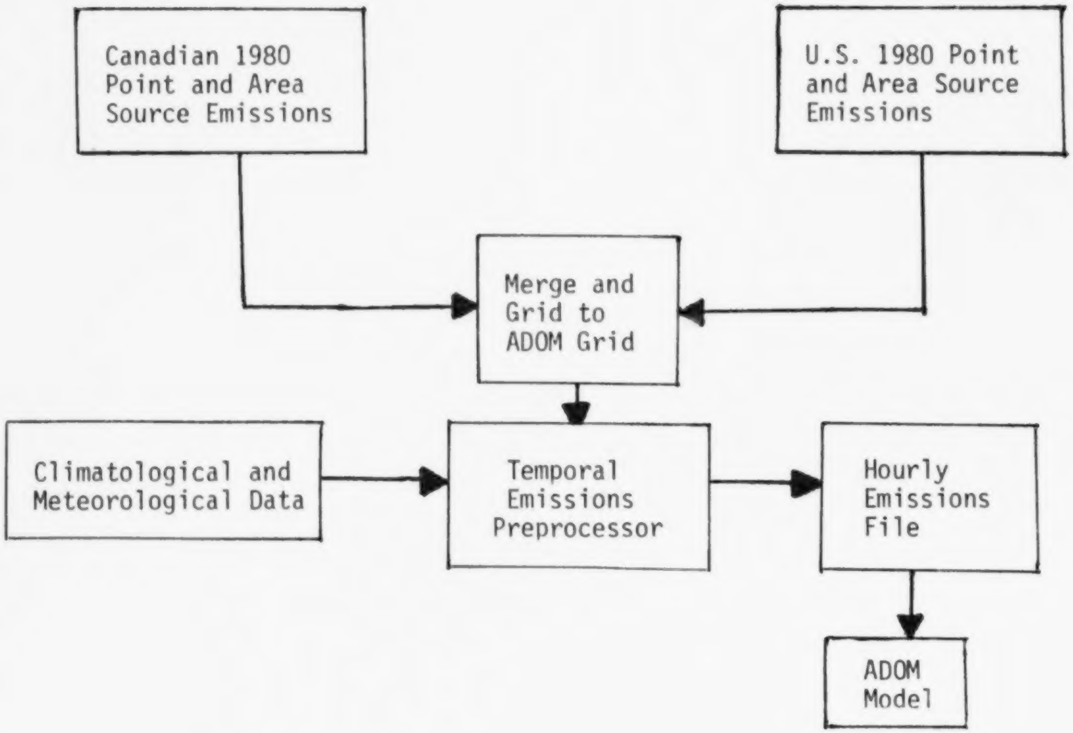

Canadian 1980 Point and Area Source Emissions
U.S. 1980 Point and Area Source Emissions
Merge and Grid to ADOM Grid
Climatological and Meteorological Data
Temporal Emissions Preprocessor
Hourly Emissions File
ADOM Model

Figure 1: Emissions Processing for the ADOM Model

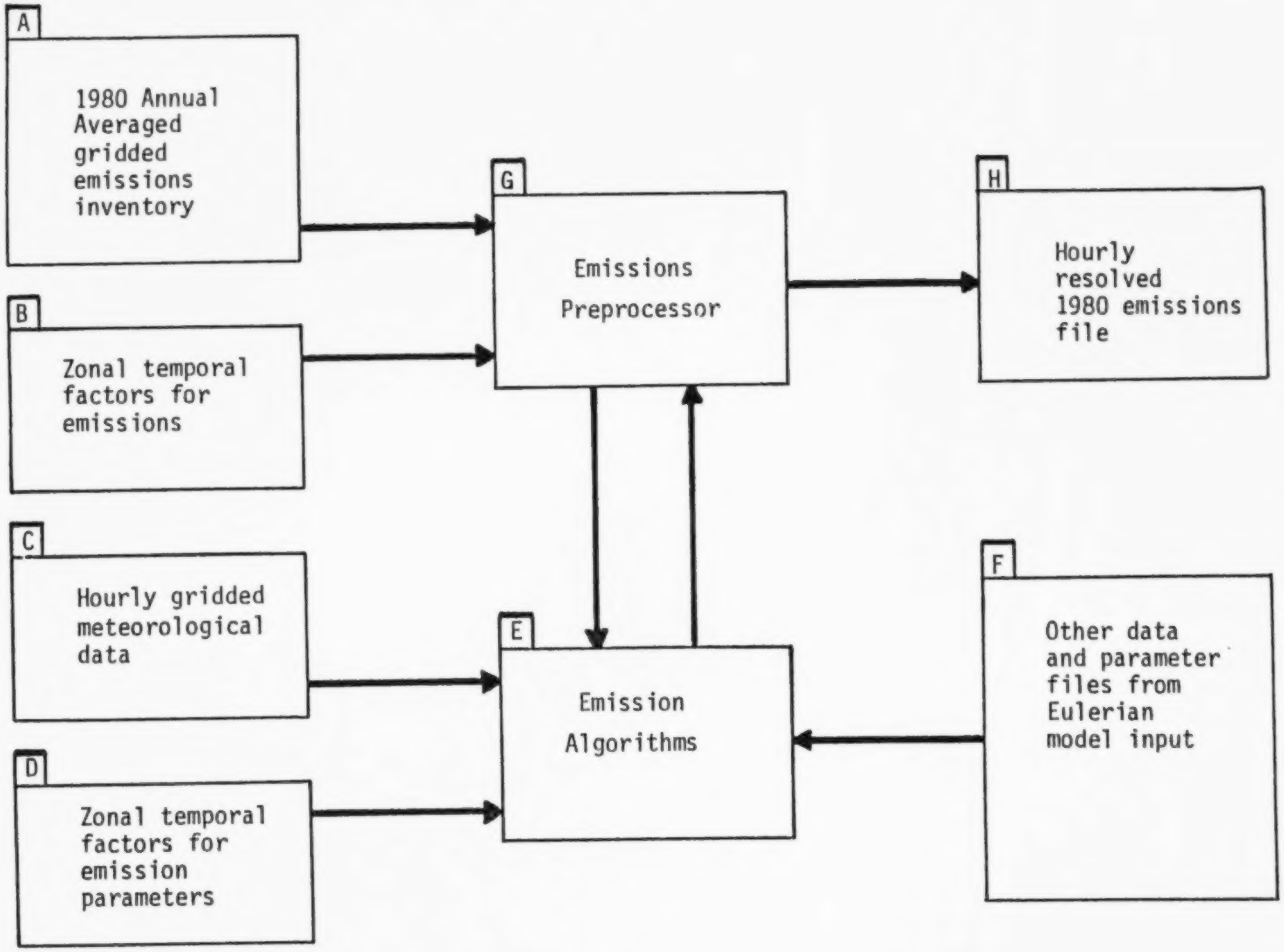
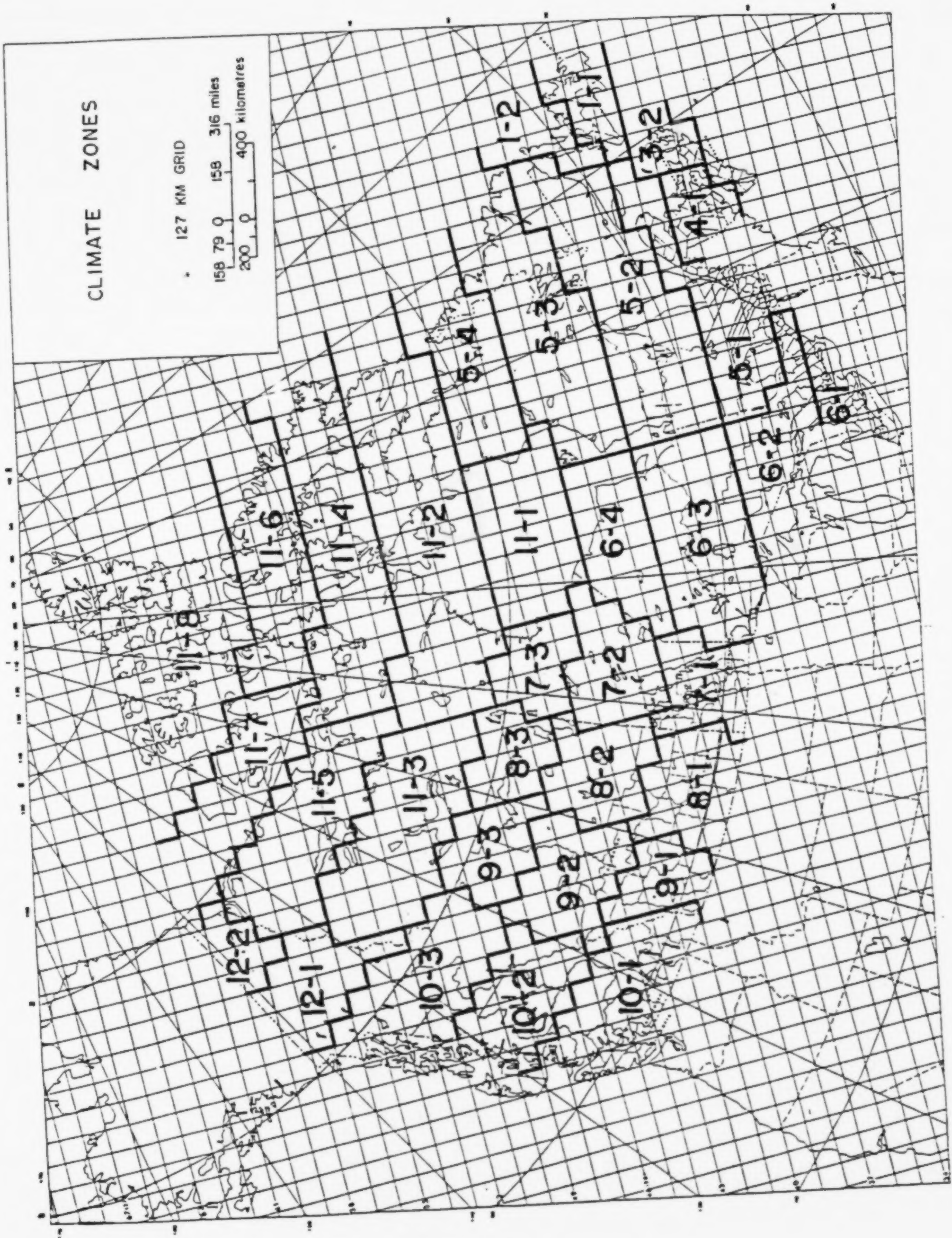

A
1980 Annual Averaged gridded emissions inventory
B
Zonal temporal factors for emissions
C
Hourly gridded meteorological data
D
Zonal temporal factors for emission parameters
G
Emissions Preprocessor
E
Emission Algorithms
H
Hourly resolved 1980 emissions file
F
Other data and parameter files from Eulerian model input

Figure 2: Computation of temporally resolved emissions for the ADOM Model.


CLIMATE ZONES
127 KM GRID
158 79 0 158 316 miles
200 0 400 kilometres
1-1
1-2
3-2
4-1
5-1
5-2
5-3
5-4
6-1
6-2
6-3
6-4
7-1
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10-2
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12-1
12-2

Figure 3

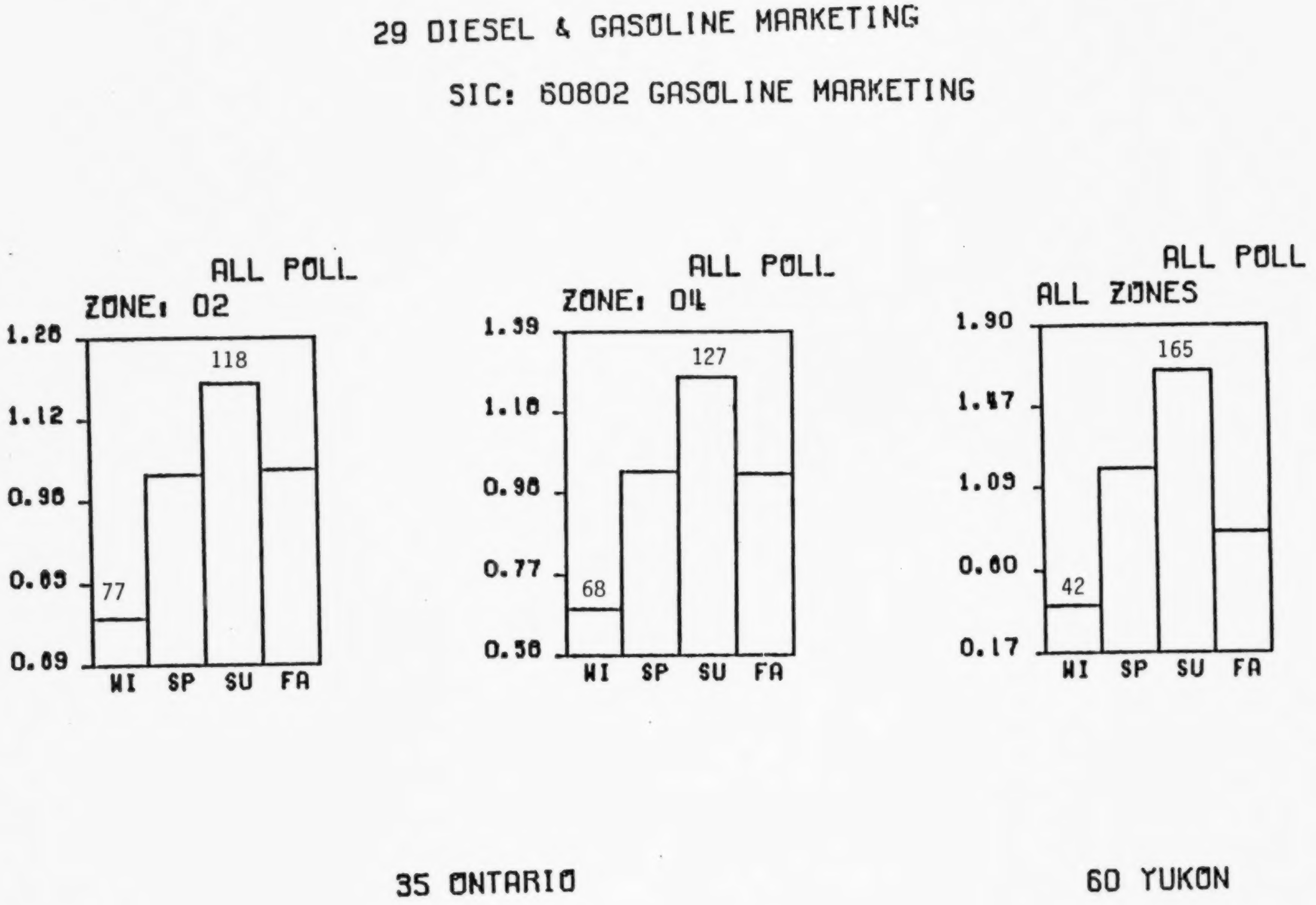
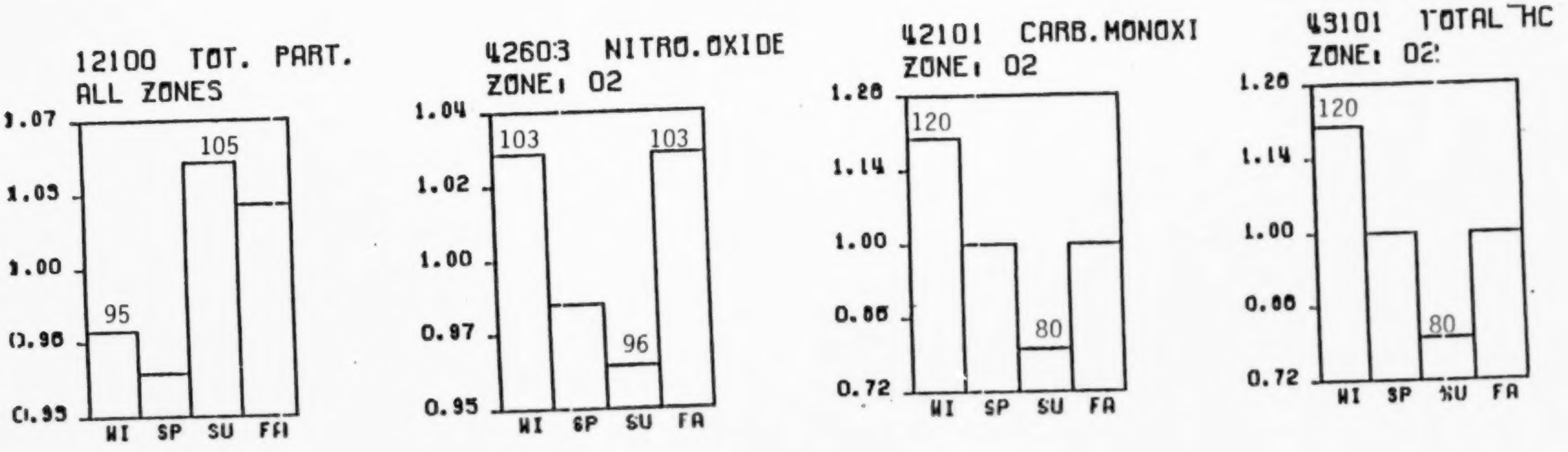
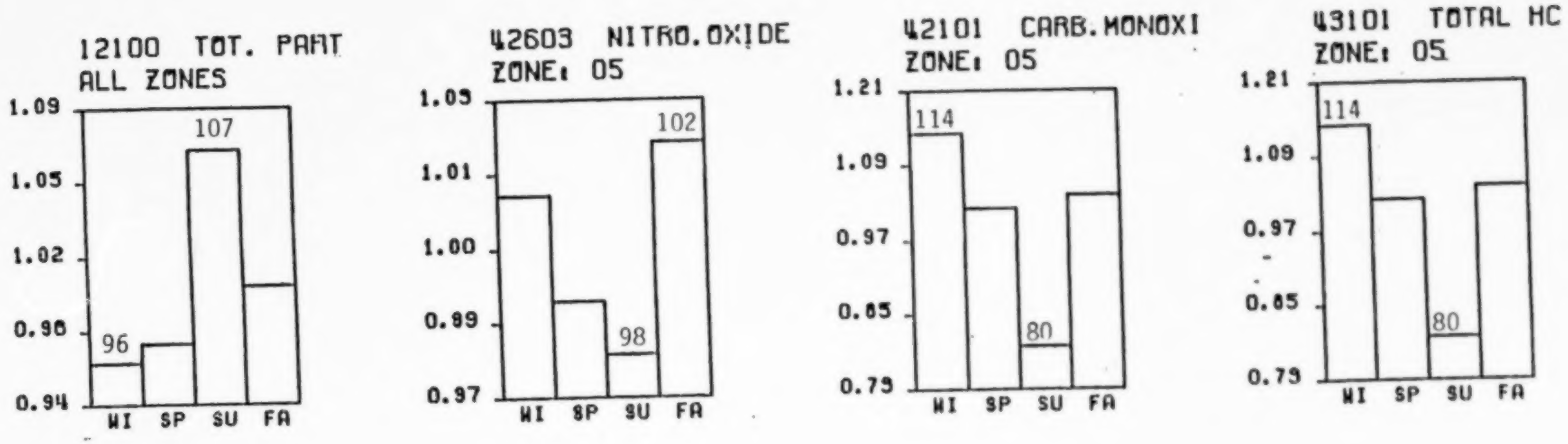


Figure 4: Seasonal temporal factors adjusted for Climate, Diesel and Gasoline Marketing.

01 GASOLINE POWERED MOTOR VEHICLE
SIC: 51000 AUTOMOBILES



35 ONTARIO



60 YUKON

Figure 5: Seasonal temporal factors adjusted for Climate; Gas Powered Motor Vehicles.

HISTORICAL ANALYSIS OF SULPHUR DIOXIDE AND NITROGEN OXIDES EMISSIONS IN CANADA

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Presented At:

Second Annual Acid Deposition Emission Inventory Symposium
Charleston, South Carolina
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ABSTRACT

Anthropogenic sulphur dioxide and nitrogen oxides emissions in Canada were calculated biennially from 1970 to 1980. For the most part, emissions were based on national statistics covering the production, distribution and consumption of goods and services and on estimates of appropriate emission factors.

The trend in the emisisons of sulphur dioxide has been steadily downward in the last decade. Traditionally the largest source of sulphur dioxide has been the non-ferrous smelting sector where emissions have decreased by 44 percent. Other major sources are the generation of thermal power from which emissions have been rising and the combustion of fuels in industrial, residential and commercial applications where a downward trend in emissions was observed. The emissions of nitrogen oxides come predominantly from transportation and the combustion of fuels, including thermal power generation, which account for about 60 and 35 percent respectively of total emissions. This ratio has remained relatively constant during the study period.

1. INTRODUCTION

The current concern with the long range transport of air pollutants (LRTAP) is with sulphur dioxide (SO_2) and nitrogen oxides (NO_x) which give rise to what has become known as "acid rain" or, more appropriately, acid deposition. This phenomenon occurs because prolonged atmospheric transport of sulphur and nitrogen oxides provides sufficient time for these compounds to undergo chemical and physical transformations into acidic compounds, which are subsequently deposited either in wet or dry form at locations distant from the originating sources of pollution. This acidic deposition causes acidification of sensitive freshwater lakes and soils, as has become evident in parts of Eastern North America, adversely affecting various ecosystems.

This paper summarizes the results of a retrospective emissions inventory for Canada of SO_2 and NO_x from 1970 to 1980. The inventory of acid rain precursor pollutants is presented nationally and regionally by major source sectors and categories. This information is important in understanding the relationships between releases of pollutants to the atmosphere and potential environmental effects and in the assessment of control options.

The source categories included in the analysis are industrial processes, fuel combustion from stationary sources, transportation and all other important anthropogenic sources. The emissions are calculated from statistics covering fuel consumption and industrial output by using consistent methodologies within each sector.

2. DISCUSSION

2.1 Methodology

The emission trends inventory was compiled on a sector level by province. The emission estimates were then aggregated to give provincial (regional) and national emissions. The calculation procedure by sector can generally be expressed as:

$$\boxed{\text{Emission} = \text{Activity Level} \times \text{Emission Factor}}$$

In order to provide a common basis for comparison for the period 1970-1980 it was necessary that each sector be treated consistently in terms of the statistical data on production, fuel consumption, etc. and using the most up-to-date uncontrolled emission factors as well as the abatement technology utilized by industry for each year of the analysis.

Most of the industrial production and fuel consumption data were derived from national statistics compiled and maintained by Statistics Canada (SC) and the Department of Energy, Mines and Resources (EMR). Emission factors were obtained primarily from the latest U.S. EPA AP-42 publication and emission control information was gleaned from the literature and determined from surveys carried out by the Department of the Environment (DOE). Average sulphur (and ash) contents of coal were derived from EMR surveys and for liquid fuels, from information supplied to DOE by fuel producers as a regulatory requirement. For the non-ferrous smelting sector, historical SO_2 emission data provided by individual companies was judged the most representative and was used for this analysis.

Transportation emissions of SO_2 and NO_x were determined in three ways: vehicle registration, fuel consumption and use statistics such as take-off and landing cycles or calls at port. Vehicle registrations were used to calculate emissions from automobiles and trucks (road vehicles); the emission factors were developed using software based on EPA'S Mobile 2.

Fuel consumption data were used to calculate the emissions from railroads as well as off-road diesel and gasoline powered vehicles. Motive fuels used in railroads were: coal, heavy and light fuel oil and diesel.

Average emission rates by engine type were used to calculate the emissions for aircraft (landing and take-off cycle) and marine (calls at port) sectors. The numbers of calls at port and LTO (landing and take-off) cycles were obtained from Statistics Canada.

2.1.1 Sulphur Dioxide

Sulphur dioxide emissions come primarily from two main source categories, industrial processes and stationary fuel combustion. The trend has been steadily downward, total national emissions going from 6.7 million tonnes in 1970 to 4.6 million tonnes in 1980 (Figure 1). Since 1970 SO_2 emissions from non-ferrous smelters, the largest emitting sector, have decreased by 44%, this includes the emissions from primary copper, nickel, lead, zinc and aluminum. As shown in Figure 2 the industrial contribution to total SO_2 emissions decreased from 76% in 1970 to 67% in 1980. The stationary fuel combustion contribution to total SO_2 emissions rose from 23% in 1970 to 30% in 1980, however in absolute terms emissions decreased by 11%. Residential, commercial and industrial (non-utility) fuel combustion SO_2 emissions decreased 40% while utility SO_2 emissions increased 52%.

The decrease in the non-ferrous smelting sector can be attributed to process improvements, increased pyrrhotite rejection at some operations and the expansion or addition of acid plants at others. The decreases in the non-utility fuel combustion emissions can be attributed to reduction of the sulphur content of fuels, the decline in the use of coal for industrial and residential use as well as a shift to cleaner fuels like natural gas and light oil.

The power utilities used additional low sulphur coal, introduced coal blending and coal washing and shifted to alternate electric power generation technologies. These measures served to limit the increase of sulphur dioxide emissions to 52%, the total power generation from coal-fired plants increased by 83% during this time.

2.1.2 Nitrogen Oxides

Nitrogen oxides predominantly come from the transportation sectors and stationary fuel combustion categories. The 30% increase in national emissions during the period 1970-1980 (Figure 3) generally reflects the increase in energy demand or fuel consumed throughout the decade. The relative amounts, as seen from Figure 4, contributed by each category to total emissions has remained essentially the same. Nitrogen oxides from stationary fuel combustion were largely uncontrolled.

In transportation, there was an increase in NO_x emissions from light-duty vehicles from 1970 to 1980. The enactment of light-duty vehicle regulations in the early seventies had a moderating effect on this increase. As the older uncontrolled vehicles are removed from the total fleet the "per vehicle" emissions decreased substantially however this trend is offset by the increasing vehicle population. The increase in transportation emissions was 36% with the largest percentage increases occurring in uncontrolled heavy duty vehicles such as trucks, off-road diesel engines, aircraft and marine equipment.

The 20% increase in nitrogen oxide emissions from stationary fuel combustion was incurred mainly because of a 60% increase in utility emissions. Despite a growing population, non-utility fuel combustion emissions were unchanged from 1970 to 1980; this reflects the change in fuel use patterns to natural gas and electricity and energy conservation practices. During the seventies Canadian utilities heavily promoted electricity for industrial and home use while government grant programs were instrumental in the rapid expansion and improvement of the gas pipeline distribution systems especially in central and eastern Canada.

3. CONCLUSION

National emission trends of sulphur dioxide and nitrogen oxides have been presented for the period 1970 to 1980. The emissions of sulphur dioxide have steadily declined with the major decrease occurring in the non-ferrous smelting sector. Nitrogen oxides have increased reflecting the general increase in energy demand, the contribution from transportation and stationary fuel combustion having increased at an equivalent rate.

Due to the limitations inherent in a retrospective emissions inventory exercise it should be stressed that the data are more suited to the study of emission trends. If comparisons to other studies are made, a prudent basis for comparison would be on the degree of change in the emissions.

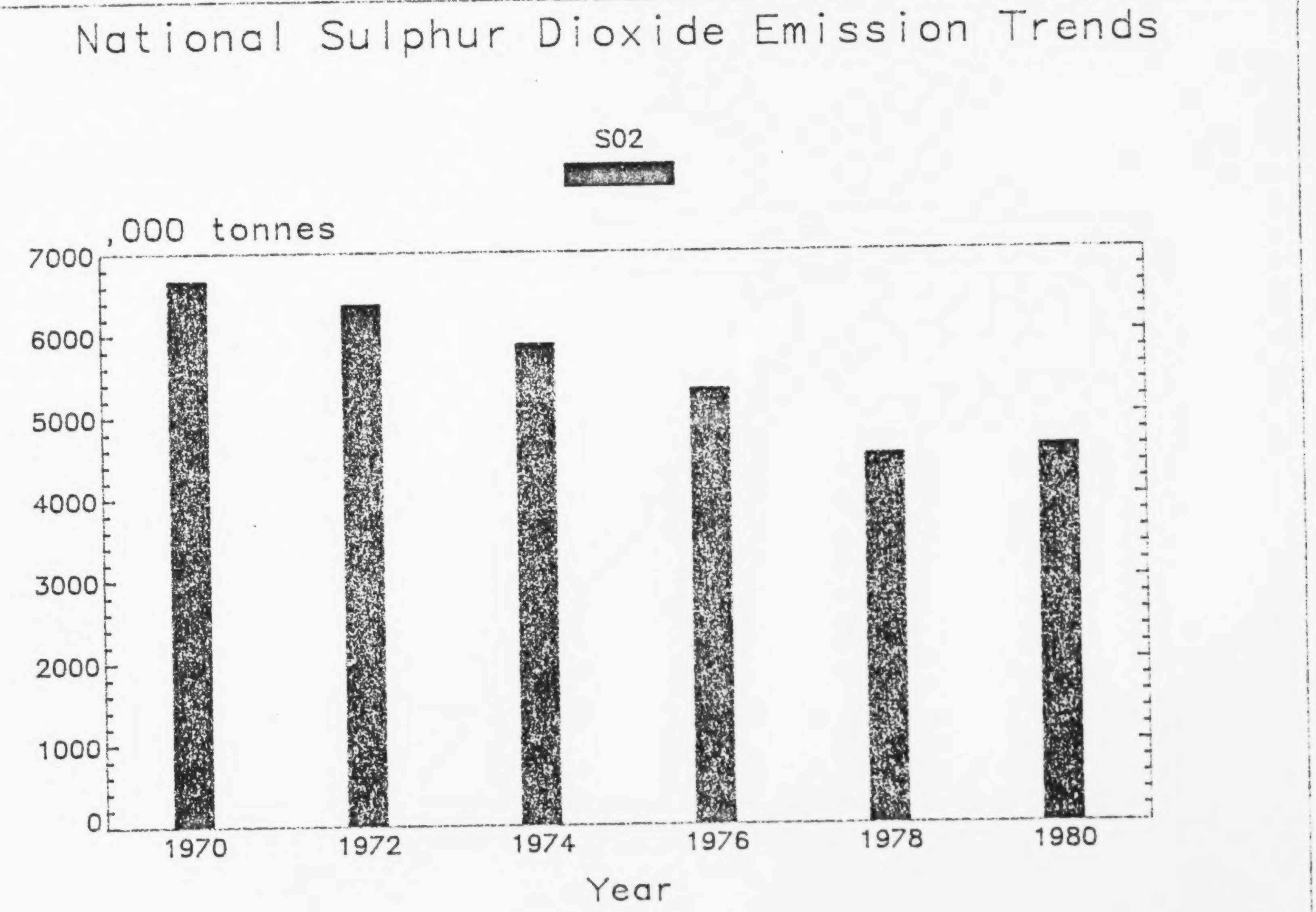
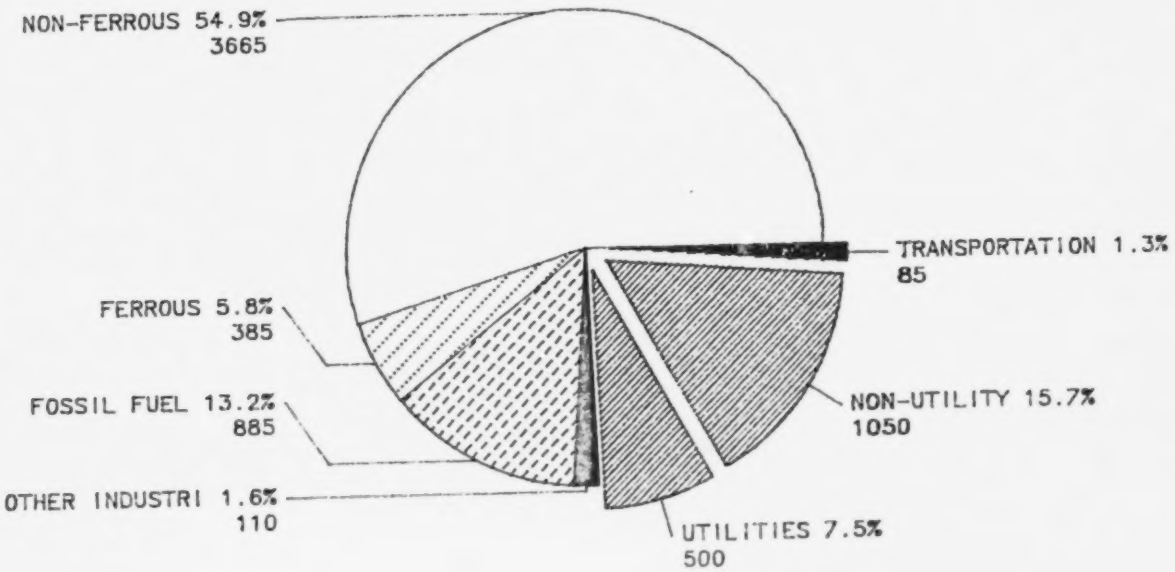

National Sulphur Dioxide Emission Trends
S02
,000 tonnes
7000
6000
5000
4000
3000
2000
1000
0
1970
1972
1974
1976
1978
1980
Year

Figure 1

DISTRIBUTION OF SO2 EMISSIONS
1970



Kilotonnes

DISTRIBUTION OF SO2 EMISSIONS
1980

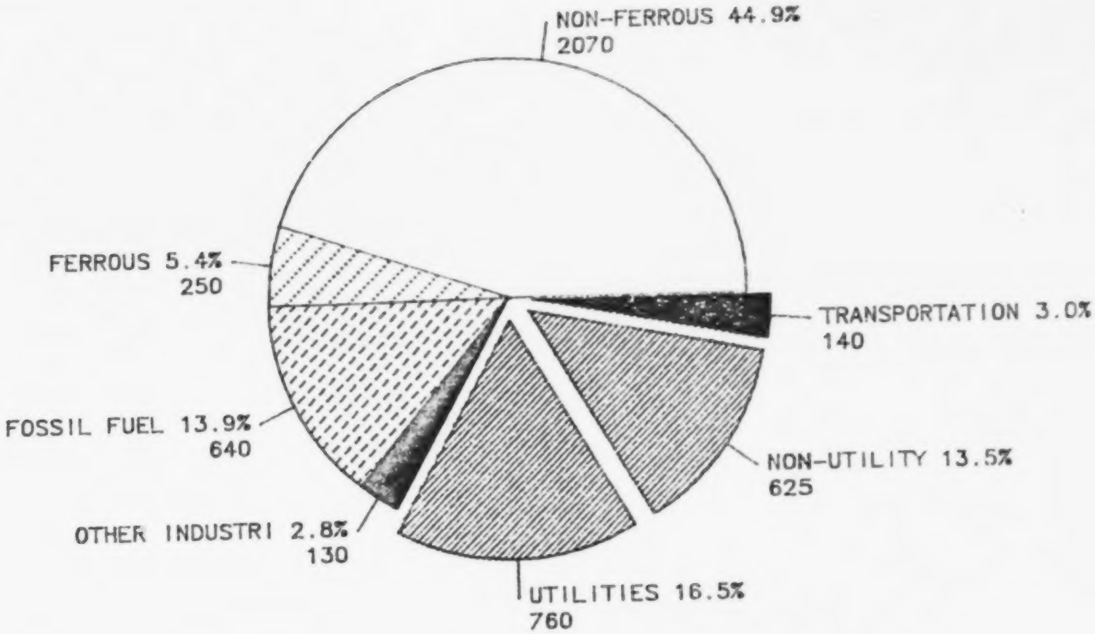


figure 2

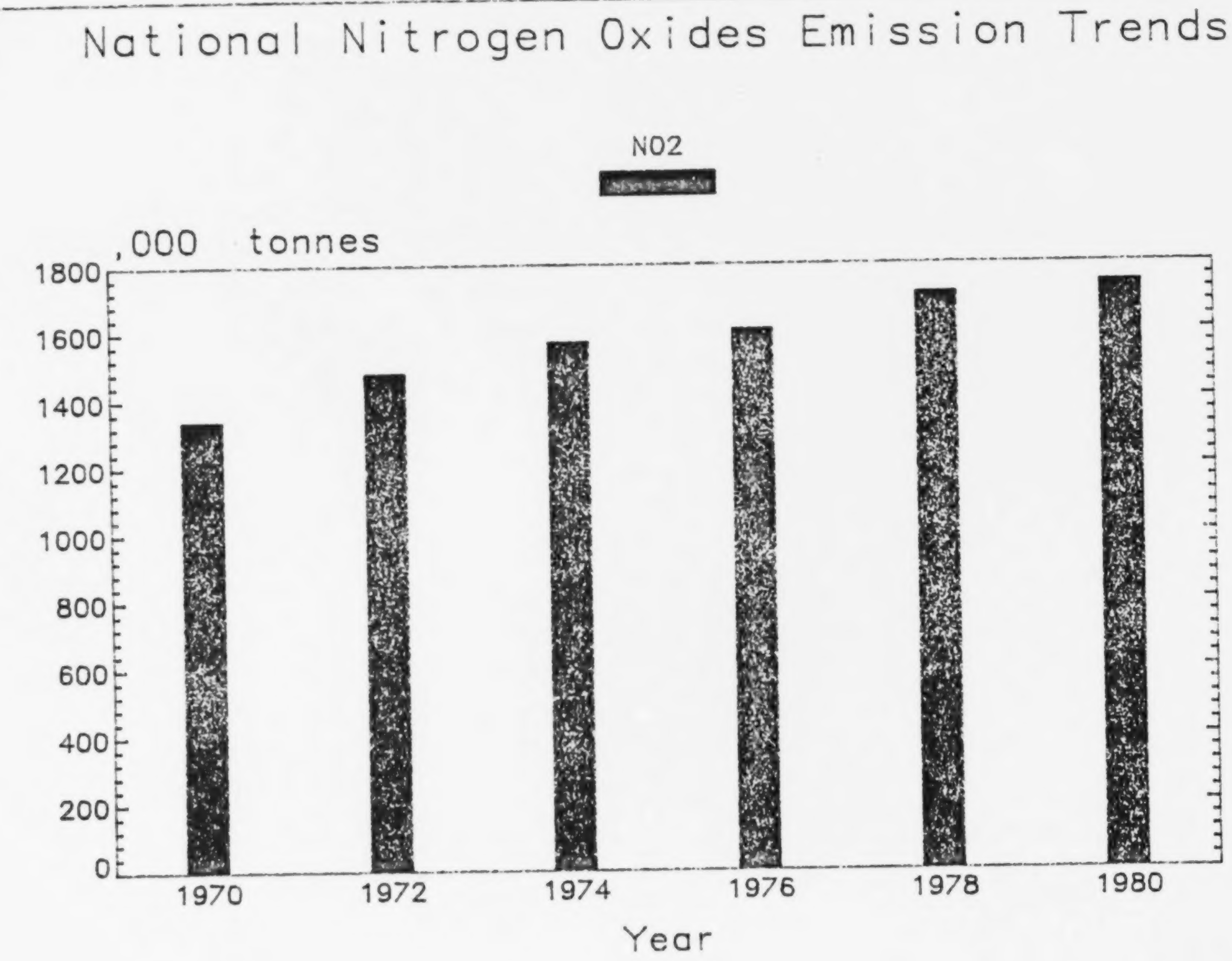
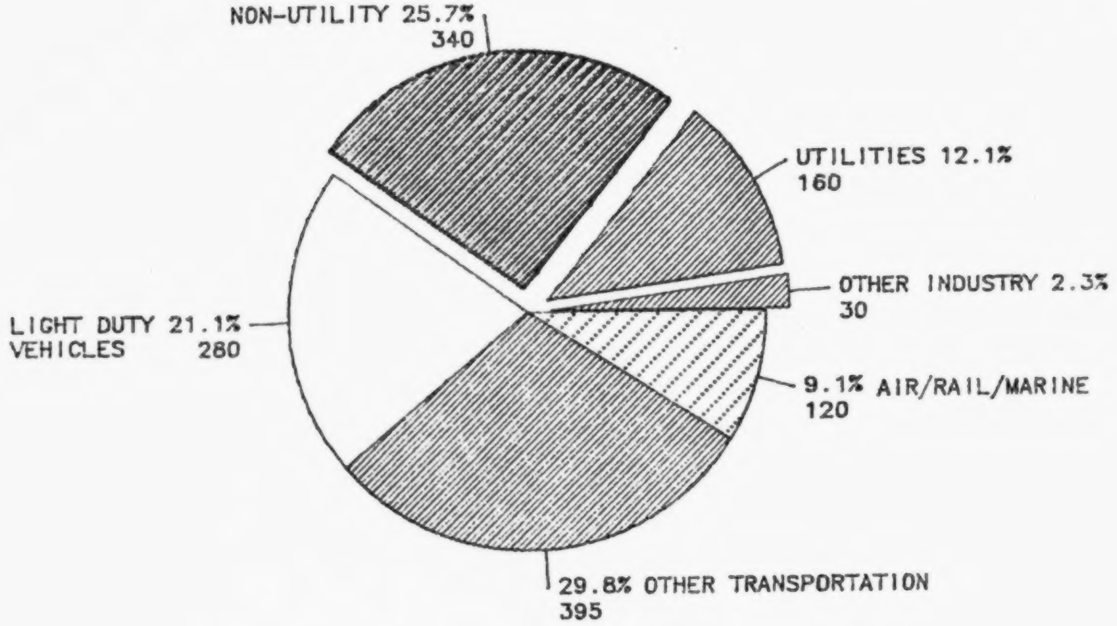

National Nitrogen Oxides Emission Trends
NO2
,000 tonnes
1800
1600
1400
1200
1000
800
600
400
200
0
1970
1972
1974
1976
1978
1980
Year

Figure 3

DISTRIBUTION OF NOX EMISSIONS

1970



Kilotonnes

1980

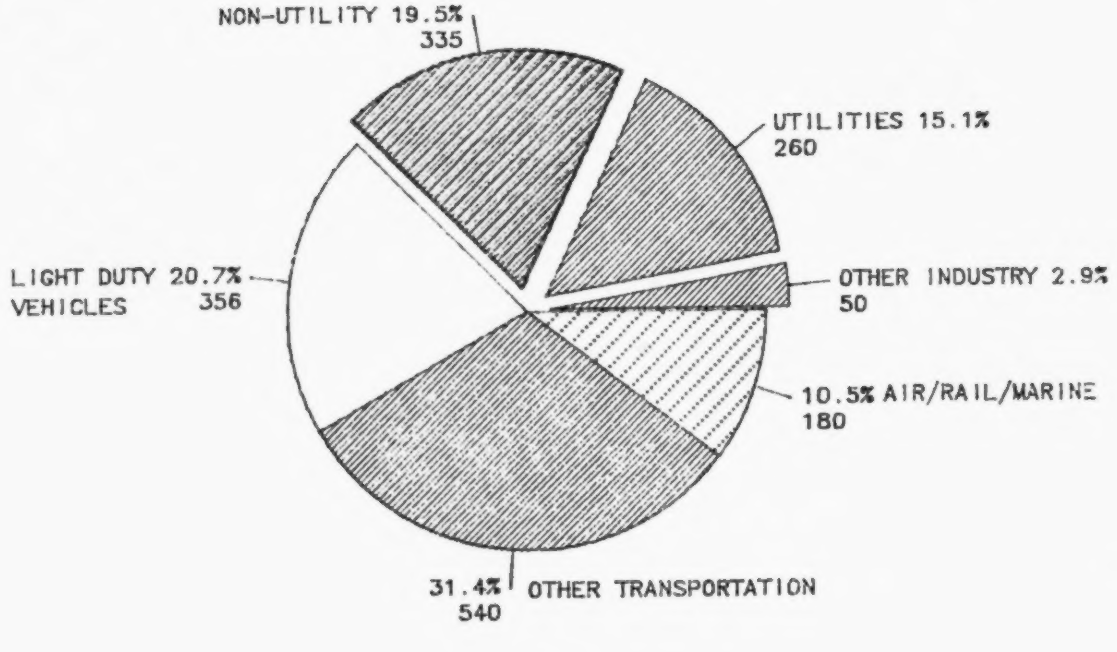


FIGURE 4

UNITED KINGDOM EMISSION INVENTORIES

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Presented at:

Second Annual Acid Deposition Emission Inventory Symposium
Charleston, South Carolina
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UNITED KINGDOM EMISSION INVENTORIES

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ABSTRACT

Warren Spring Laboratory (WSL) is an environmental/industrial research establishment of the United Kingdom Department of Trade and Industry. WSL's Air Pollution division is a major center for research on the physical and chemical aspects of air pollution in the United Kingdom and covers emission, dispersion and ambient air quality. National emission estimates for SO_2, NO_x, CO and VOC have been made for several years. A new project is now in progress to produce spatially disaggregated emission inventories for the United Kingdom. Estimates of SO_2 have been made and work is proceeding to cover other pollutants, initially NO_x and VOC. The disaggregated inventories will be based on a relational data base which, in addition to air quality information, will contain emission information on large plants and estimates of area emissions mainly on a 20 x 20 km grid basis using emission information and surrogate statistics to cover smaller emission sources. Emissions from fuel combustion as well as from industrial processes and evaporative emissions are included using, where available, emissions factors based on emission measurements made under United Kingdom conditions by WSL and others. The emissions data base will be used by WSL primarily in numerical dispersion modeling of urban and longer range transport, chemical reactions and depositions, as well as assisting in the assessment and interpretation of the data from the national United Kingdom monitoring networks for acid deposition, smoke, SO_2, NO_x, and ozone, coordinated by WSL.

UNITED KINGDOM EMISSION INVENTORIES

INTRODUCTION

Warren Spring Laboratory (WSL) is a research establishment of the United Kingdom Department of Trade and Industry whose interests cover environmental and industrial fields. The Air Pollution division is a major center for research on the physical and chemical aspects of air pollution in the United Kingdom and covers emission, dispersion and ambient air quality. Current research topics include the measurement of combustion and process emissions from mobile and stationary sources, modeling of air pollution, and the operation and coordination of monitoring networks measuring smoke and SO_2, acid deposition, NO_x, ozone, hydrocarbons and suspended particulates including lead. New projects are the construction of a national automated network of NO_x and ozone measurement sites, the development of spatially disaggregated emission inventories and the creation of a new computer system based on a relational data base. The data base will include information held by WSL on emissions and related statistics, measurements of air quality and deposition, meteorological data and other relevant information.

The different monitoring networks are designed for a variety of purposes. The smoke and sulphur dioxide monitoring networks were created in 1961. They provide daily measurements and are voluntarily operated sites coordinated by WSL. The aim was to provide baseline information about the distribution and trends of smoke and SO_2 in the United Kingdom. This was reorganized in 1982 into a number of networks with separate aims. Continuous monitoring of NO, NO_2, CO, O_3 and SO_2 also is undertaken by WSL. Sites representative of urban and rural situations have been monitored for up to 10 years. The results of these surveys are published and summaries are given in the DOE Digest of Environmental Protection and Water Statistics. The continuous monitoring network is being extended with the construction of a network of fully automatic NO_x/O_3 monitoring stations. There is also a national network of sites measuring pollution concentration in rain. A network of remote rural sites collects samples of precipitation and these are analyzed for a variety of ions. These results plus sulphur measurements form the United Kingdom input to the United Nations Economic Commission for Europe's European Monitoring and Evaluation Program investigating the long-range transport of air pollutants.

EMISSION INVENTORIES

National Aggregates

For several years WSL has estimated national emissions of SO_2, NO_x, CO and hydrocarbons in the United Kingdom. These estimates are mainly based on emissions from fuel combustion, but they do include estimates of emissions from other sources. Originally, the emission factors used relied heavily on American data but now most are based on measurements made under United Kingdom conditions by WSL and others. These estimates were published in the Department of the Environment Digest of Environmental Protection and Water Statistics (Reference 1).

Emissions of SO_2 since 1960 are shown in Figure 1. The emissions were about 6 million tonnes during the 1960s, but have not fallen by about 40 percent to 3.54 million tonnes in 1983. This has been due to the large reduction in low and medium level sources and has resulted in a reduction in average urban levels of SO_2 of over 70 percent. This reduction is still continuing and the total emissions have fallen by nearly 25 percent since 1980.

The estimates of emissions for 1983 are shown in Table 1. These figures show clearly the most significant sources of each pollutant. Over 70 percent of SO_2 is emitted from power stations and most of the remainder from industrial sources. The largest sources of NO_x are power stations and road transport both emitting about 40 percent of the total. Road vehicles also account for nearly 84 percent of the CO and are a significant source of non-methane hydrocarbons emitting a similar quantity to solvent evaporation and industrial processes.

Recent measurements, made on 72 commercial and industrial boilers in Scotland (Walker et al., Reference 2), with sizes between 430 and 14,200 kW, have resulted in the adoption of new emission factors for NO_x, CO and VOC.

Comparison of the factors with the previously used factors is complicated because the study used different size ranges for the boilers. However, the factors used to calculate the national emissions have been altered as shown in Table 2.

Many of the factors are only altered slightly, but for some there are more substantial differences. As a result of these changes, the estimate of emissions from commercial and industrial fuel consumption has changed. A comparison is shown in Table 3 for the year 1982.

Until 1983, vehicle emissions were estimated on the same basis as emissions from stationary sources by applying an emission factor to the total fuel consumed. However, as emissions from vehicles are related to the speed of the vehicle and the manner in which it is driven as well as the amount of fuel consumed, this method took no account of varying patterns of road use. Measurements on the road, rather than on a dynamometer, were made by WSL of emissions from motor vehicles at a range of speeds so that the variation of emission with speed could be determined (Potter and Savage, Reference 3). Emission factors for pollutant emission per kilometer driven at each speed could then be estimated. The sample of cars tested was chosen to reflect the makeup of the United Kingdom car fleet. Road traffic statistics give the number of vehicle-kilometers driven on different types of roads and the average speeds of different categories of vehicles. Measurements of vehicle speeds in the United Kingdom indicate that the distribution of vehicle speed about the mean is approximately gaussian with a standard deviation of one-sixth of the mean. This enables a speed distribution for a particular type of road to be calculated and then using the measured speed-related emission factors, the emissions can be estimated (Reference 4). Emissions estimated using this method have the advantage that they reflect the types of roads and driving patterns and are sensitive to any changes in them. A comparison of the results obtained using the single emission factor method and the new speed-related method is given in Table 4.

Data Base

WSL has recently installed a new GEC 63/30 computer running the AT&T UNIX System 5 operating system. It has 8 M bytes of random access memory, over 1 G byte of backing store and is accessed through an open-system local area network which is also connected to other computers and services. A relational data base management system, MISTRESS, has also been purchased and will be used to store the national air pollution data base.

The data base will be central to the work of the air pollution division of WSL. It will store all the information relating to air pollution: ambient air monitoring, acid deposition measurements and meteorological data, was well as emission inventories. The system is illustrated in Figure 2. Data will be processed into the data base and will be available for comparison with other data or with model results. The emissions data will be collected in separate data base files along with surrogate statistics. Only completed and validated inventories will be stored in the main data base and be available for general interrogation and input to modeling programs. The data base will be linked to statistical and graphical packages as well as report writing facilities. The system, which should be fully operational in the first half of 1986, will provide an integrated and comprehensive package of retrieval, presentational and analytical routines.

Spatially Disaggregated Inventories

The estimation of spatially disaggregated inventories for the United Kingdom is a new project started this year. The inventories are to be prepared on a 20 x 20 km grid for the whole United Kingdom but smaller grids will be used for some areas.

A few local authorities have compiled local, spatially disaggregated inventories and these are being collected by WSL. They are mainly compiled from questionnaires to local firms and estimates of local fuel consumption in various sectors. These can be used as direct input to the national inventories but will also be used to help validate the disaggregation procedures adopted to provide information for the United Kingdom as a whole.

The Greater London Council has compiled an inventory for London in 1976 and is in the process of updating this. Sheffield and Coventry have inventories for SO_2 in 1983, compiled from fuel use information.

There is no comprehensive national program for the routine measurement of emissions in the United Kingdom, so that there are very few actual emission measurements available for inclusion in an emission inventory. Emissions, therefore, must be estimated from other information available about individual sources or by using surrogate statistics. Where appropriate, emission sources will be stored as point sources within the data base, otherwise emissions will be aggregated on an area basis. The following approaches have been adopted by WSL for the production of the spatially-disaggregated inventories for the United Kingdom.

Power Stations: most of these are publicly owned and information is available on type, age, power output and efficiency of individual stations. Pollutant emission factors can, therefore, be applied to the appropriate statistic, fuel consumption or power output, in order to estimate emissions from each station. To date, it has been assumed that 10 percent of the sulphur in coal is retained in the ash at each station; recent evidence suggests that this may be a high estimate of retention and investigations are under way to determine a more realistic figure.

Other industrial sources: directly relevant information on these are not so readily available. Contact is being established with the relevant industry associations with a view to obtaining more information on individual large plant on a voluntary basis and questionnaires are being used to obtain such information in a consistent and readily usable form. To date, emissions from large emission source categories, where available, have been estimated from emission factors and national throughput disaggregated in proportion to plant capacity to provide estimates of point source emissions.

Emissions from other stationary sources including smaller industrial source categories, commerce, and housing will be treated as area sources. These emissions will be estimated from spatially disaggregated statistics on fuel deliveries or from nationally aggregated statistics, for example production, output or national fuel consumption and then disaggregated using an appropriate surrogate statistic (e.g., population or employment).

A statistical data base, called PINGAR, is available for petroleum deliveries into 10 km squares for the whole United Kingdom with densely populated areas covered at a 1 km resolution. These statistics cover different fuels and users and provide a basis for estimation of emissions from all stationary petroleum consumption. While this will not directly provide data on individual point sources, it will locate accurately consumption within the correct grid square. Other surrogate statistics are not usually available on a grid basis; an exception is census data for Scotland. Other statistics which can be used are population and household density from census returns, land use, employment and agriculture. These are provided for various areas (e.g., counties) and have to be transposed to a grid before they can be used. Spatially disaggregated domestic emissions will be estimated from regional coal consumption and population.

Motor vehicles contribute over 40 percent of the NO_x and VOC emissions so considerable effort is necessary to account adequately for these sources. In London, vehicle counts have been made so that total vehicle kilometers are available for each borough for motorways and other roads and for peak and off-peak hours together with average speeds, so it is possible to use the speed-related emission factors to estimate emissions in each borough. Outside London, there is less information available on traffic on a spatially resolved basis, but the Department of Transportation does produce statistics on vehicle kilometers for each county in the United Kingdom. Another source of information is the 1981 Census which provides statistics on the distribution of cars in the country. This will be used to disaggregate the county estimates.

The emission inventory data base will contain details of individual point sources - source category, location, height, number and size of stacks, thermal input, abatement details (if present), fuel consumption and type, period covered by the information, etc., as well as emission estimates. It will, therefore, be possible to interrogate the data base using a number of search criteria. For example, emission estimates could be computer-mapped to show the spatial distribution or used as input to a predictive model. It will also be possible to use the system to convert the emission estimates from one spatial-grid system to another.

Sulphur Dioxide

Using the above mentioned approaches, an emission inventory for SO_2 on a 20 x 20 km grid, based on the United Kingdom's Ordinance Survey grid, has been produced for 1983 and is shown in a graphical form in Figure 3.

Nitrogen Oxides

The production of a disaggregated inventory for NO_x involved a similar approach to that for SO_2 with the major difference being that more effort was required to account for emissions from vehicle sources as motor vehicles contribute about 40 percent of the emissions. The accuracy of the disaggregated inventory will be dependent on the method used to distribute these emissions across the grid cells. Using the method described above, a first estimate has been made and this could be further refined with the use of more appropriate traffic statistics.

Other Inventories

The most important of these will be the hydrocarbon inventory. The two main sources are vehicles, which are dealt with as above, and evaporative and process emissions. The latter will be difficult to quantify accurately in a spatially disaggregated inventory due to both the lack of individual plant or area production/consumption data and reliable emission factors for individual hydrocarbons. For the foreseeable future, surrogate statistics (e.g., industrial floorspace or employment in particular industrial sectors) and bulk hydrocarbon emission factors will continue to be used.

Active consideration is being given to other pollutants such as NH_3. This will require further study to determine the major sources and the best statistics that are available to describe them.

Another long-term aim is to provide temporal disaggregation of emissions. Relevant information is being collected, but such refinements will take second priority to the annual inventories.

CONCLUSIONS

Spatially disaggregated emission inventories are being produced for a variety of atmospheric pollutants. In the United Kingdom, surrogate statistics have to be used to overcome the scarcity of emission measurements. Future work will involve the establishment of inventories of other pollutants and the estimation of temporally disaggregated inventories, as well as the refinement of existing inventories.

This work will be integrated via the new data base system with monitoring and modeling of air pollution in the United Kingdom and Europe to improve our understanding of the dispersion and deposition of atmospheric pollutants.

REFERENCES

1. Digest of Environmental Protection and Water Statistics No. 7 (1984), HMSO, London, United Kingdom.

2. Walker, D.S., Galbraith, R., and Galbraith, J.M., Warren Spring Laboratory, Report LR 524 (1985), Survey of Nitrogen Oxides, Carbon Monoxide and Hydrocarbon Emissions from Industrial and Commercial Boilers in Scotland.

3. Potter, C.J., Savage C.A., Warren Spring Laboratory, Report LR 470 (1983), A Survey of Gaseous Pollutant Emissions from Tuned In-Service Gasoline Engined Cars over a Range of Road Operating Conditions: Executive Summary.

4. Rodgers, F.S.M., Warren Spring Laboratory, Report LR 508 (1984), A Revised Calculation of Gaseous Emissions from United Kingdom Motor Vehicles.

TABLE 1. NATIONAL EMISSIONS OF AIR POLLUTANTS 1983 (MILLION TONNES)

	SO_2		NO_x		CO		VOC	
	Mt	%	Mt	%	Mt	%	Mt	%
By Fuel:								
Coal	2.81	76	0.80	43	0.37	7	0.08	6
Solid Smokeless	0.10	3	0.04	2	0.20	4	--	--
Motor Spirit	0.02	--	0.52	28	3.99	79	0.49	39
DERV	0.03	1	0.19	10	0.25	5	0.04	3
Burning Oil	--	--	--	--	--	--	--	--
Gas Oil	0.05	2	0.06	3	0.02	--	0.01	1
Fuel Oil	0.53	14	0.09	5	0.01	--	--	--
Refinery Fuel	0.16	4	0.03	2	--	--	--	--
All Gas	--	--	0.08	4	0.01	--	--	--
By Consumer:								
Domestic	0.19	5	0.05	3	0.46	9	0.07	6
Commercial/Public Service	0.14	4	0.04	2	0.01	--	--	--
Power Stations	2.53	68	0.76	41	0.05	1	0.01	1
Refineries	0.16	4	0.03	2	--	--	--	--
Agriculture	--	--	--	--	--	--	--	--
Other Industry	0.52	17	0.18	10	0.07	1	--	--
Rail Transport	--	--	0.04	2	0.01	--	0.01	1
Road Transport	0.04	1	0.70	38	4.23	83	0.53	42
Incineration	--	--	0.01	1	0.22	4	0.04	3
Process and Evaporation	--	--	--	--	--	--	0.60	47
TOTAL	3.72		1.85		5.08		1.27	

Note: The figures are mainly based on fuel related emission factors and will underestimate the emissions from industrial processes of all but VOCs. This will only be a few percent of the total emissions. The total for hydrocarbons excludes leakage from the distribution system. This is mainly methane and amounted to about 2.2 million tonnes in 1983.

TABLE 2. COMPARISON OF "OLD" AND "NEW" COMMERCIAL AND INDUSTRIAL EMISSION FACTORS

Fuel		Emission Factor (g/kg or Gas kg/Mtherm) "Old"[a]	Emission Factor (g/kg or Gas kg/Mtherm) "New"[b]
NO_x	Coal	7.79	4.8
	Solid Smokeless	7.79	4.8
	Gas Oil[c]	2.6	2.6
	Fuel Oil	7.5	7.4
	Natural Gas	5280	5340
CO	Coal	6.64	4.1
	Solid Smokeless	6.64	4.1
	Gas Oil[c]	0.24	0.24
	Fuel Oil	0.5	0.50
	Natural Gas	247.5	250.0
VOC	Coal	0.115	0.07
	Solid Smokeless	0.115	0.07
	Gas Oil[c]	0.07	0.07
	Fuel Oil	0.06	0.06
	Natural Gas	165.0	153.0

[a] The "old" factors are based on U.S. EPA factors, but they have been updated to take into account previous measurements in the United Kingdom.

[b] The "new" factors result from a series of measurements made on United Kingdom boilers (Reference 2).

[c] The factor for Gas Oil has not been updated as too few measurements were made to provide an accurate figure, however, the measurements agreed with the "old" value rather than with the U.S. EPA factor.

TABLE 3. EMISSIONS FROM COMMERCIAL AND INDUSTRIAL FUEL CONSUMPTION 1983

	Emission (k tonnes)	
	Old[a]	New[b]
NO_x	221 (13%)[c]	187 (11%)
CO	82 (1.6%)	54 (1.1%)
VOC (non-methane)	4 (0.3%)	3.2 (0.2%)

[a] Old refers to the U.S. EPA based (but modified by United Kingdom measurements) factor.

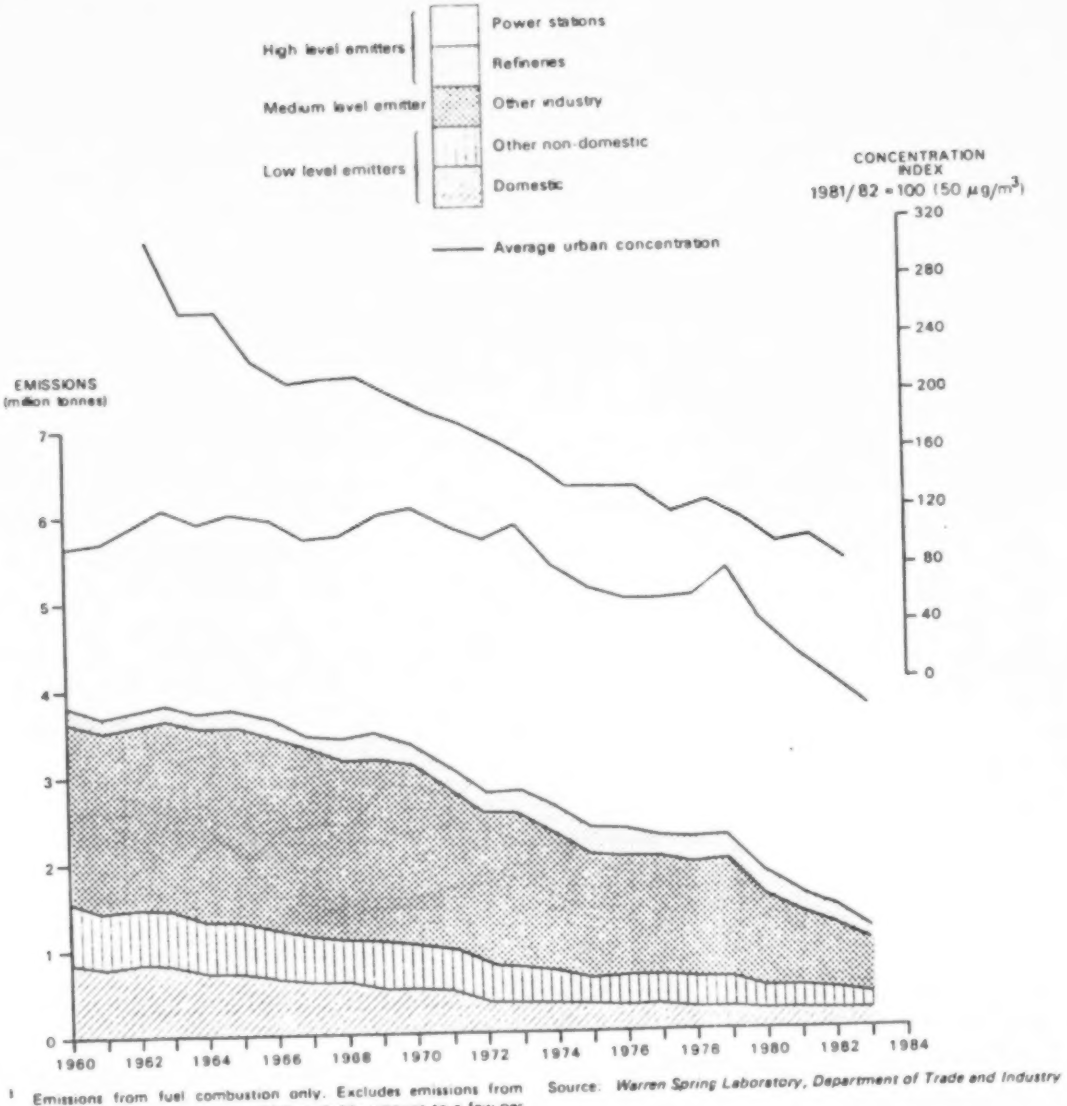
[b] New refers to the factor based on measurements made in the United Kingdom.

[c] The numbers in brackets are percentages of the national total emission.

TABLE 4. EMISSIONS FROM MOTOR CARS (1983)

	Single Emission Factor[a]	Speed Related Factors
NO_x	508	725
CO	8107	4502
VOC	538	538

[a] (k tonnes).



[1] Emissions from fuel combustion only. Excludes emissions from chemical and other processes which probably amount to a few per cent of total emissions from fuel combustion.

Source: *Warren Spring Laboratory, Department of Trade and Industry*

Total emissions of sulphur dioxide in the United Kingdom were relatively stable throughout the 1960s and early 1970s at about 6 million tonnes a year. After 1973, however, annual emissions fell to about 5 million tonnes in 1976, 1977 and 1978, to 4.0 million tonnes in 1982, and to 3.7 million tonnes in 1983. Average urban concentrations have fallen by over 70 per cent since 1962, in line with the reduction in emissions from low-level sources (see Table 2.4 and Additional Tables A1 and A3).

Figure 1. Sulphur dioxide emissions from[1] fuel combustion and average urban concentrations.

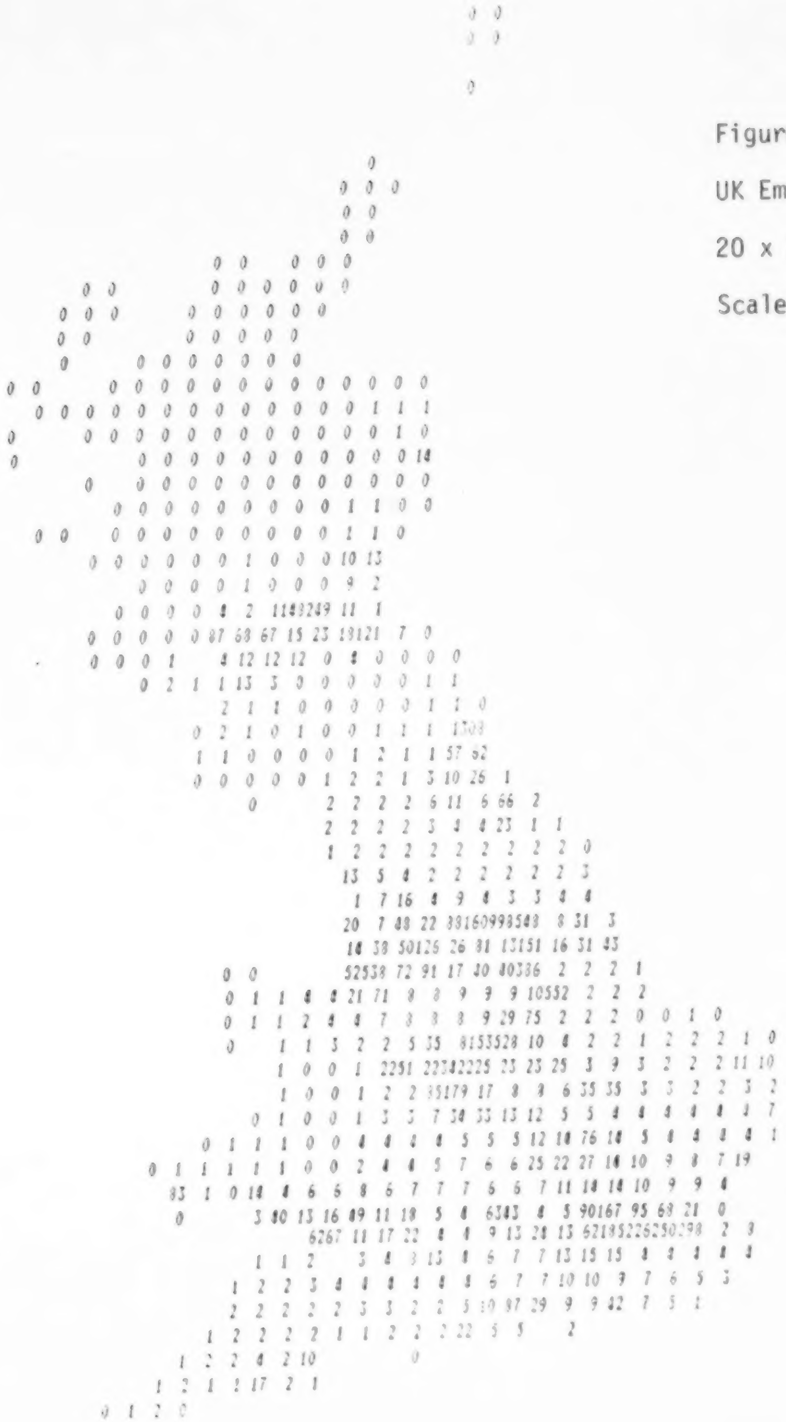


Figure 2.

UK Emissions of SO_2

20 x 20 km grid

Scale: 1=346.5 tonnes

APPENDIX

ATTENDEES

SECOND ANNUAL ACID DEPOSITION SYMPOSIUM

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